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IMPROVEMENT OF MODEL RELIABILITY IN A SOIL-PLANT SYSTEM

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SYMBOLS LIST

a	constant
a_{clean}	cleanup rate per worker [m^2 man^{-1} h^{-1}]
a_p	root absorption coefficient [mL cm^{-2} min^{-1}]
A	contaminated area under consideration [m^{-2}]
ALI	annual limit of intake for a worker [Bq y^{-1} man^{-1}]]
ALI_c	annual limit of intake for a consumer [Bq y^{-1} man^{-1}])
b	constant
C_{crop}	radionuclide concentration in a crop [kBq kg^{-1}]
C_f	radionuclide concentration in free space in root [Bq mL^{-1}]
C_i	initial radionuclide concentration in solution [Bq L^{-1}]
C_l	radionuclide concentration in soil solution [Bq L^{-1}]
C_{limit}	radionuclide concentration in soil above which cultivation should be restricted [kBq kg^{-1}]
C_s	radionuclide concentration in soil solid phase [Bq g^{-1}]
C_{safe}	safety level for residence [Bq g^{-1}]
C_{soil}	radionuclide concentration in surface soil [Bq g^{-1}]
C_x	radionuclide concentration in xylem [Bq mL^{-1}]
CDR	cation distribution ratio between soil and solution which is defined as the value of soil CEC divided by solution EC
CEC	cation exchange capacity of soil [mmol(+) $100g^{-1}$-soil]
D	dispersion coefficient [cm^2 min^{-1}]
D_{life}	additional cumulative dose that each resident will get in his life time [Sv]
D_{limit}	maximum dose limit [Sv]
D_{worker}	predicted dose to a worker [Sv]
d_s	depth of root zone soil [cm]
EC	electrical conductivity in soil solution [mS cm^{-1}]
EC_m	electrical conductivity in supernatant [mS cm^{-1}]
f_e	concentration coefficient in edible parts by their weight [g^{-1}]
f_{CR}	cancer risk due to internal radiation of 1 Sv [man Sv^{-1}]
f_{DE}	ingestion dose equivalent [Sv kBq^{-1}]

F_{conv}	conversion coefficient from radionuclide concentration in surface soil to dose equivalent per resident [Sv yr^{-1} per Bq kg^{-1}]
F'_{conv}	conversion coefficient from radionuclide concentration in surface soil to dose equivalent per worker [Sv yr^{-1} per Bq kg^{-1}]
F_s	ion flux from xylem to shoot (above ground parts) [Bq h^{-1}]
I_c	annual crop consumption by a consumer [kg y^{-1} man^{-1}]
I_d	daily intake of a crop per consumer [kg man^{-1} d^{-1}].
I_y	annual radionuclide intake by a consumer [Bq y^{-1} man^{-1}]
k_w	water absorption coefficient per unit root surface [mL cm^{-2} h^{-1}]
$K_{a/b}$	selectivity coefficient [-]
K_d	soil-solution distribution coefficient [L kg^{-1}]
$K_{d\ a}$	soil-solution distribution coefficient of ion a [L kg^{-1}]
$K_{d\ b}$	soil-solution distribution coefficient of ion b [L kg^{-1}]
K_{fl}	distribution ratio of radionuclide ions between free space and soil solution [-]
L	total root length [cm]
λ	effective removal constant [y^{-1}]
λ_c	rate of root uptake by crops [y^{-1}]
λ_d	radioactive decay constant [y^{-1}]
λ_{di}	decay constant when cultivation is restricted (= $\lambda_d + \lambda_i$) [y^{-1}]
λ_i	rate of downward migration into below subsurface soil [y^{-1}]
m_{comp}	annual cost for compensation per resident [$ man^{-1} y^{-1}]
M	total amount released from the source [kBq]
M_{comp}	total compensation cost [$]
N_c	number of consumers per farmer [man]
N_{ex}	predicted number of residents who will get more than the maximum dose limit [man]
$N_{resident}$	number of residents [man]
N_{worker}	number of workers [man]
P_{ex}	probability of getting more than the maximum dose limit [-]
P_{xf}	concentration ratio between xylem and free space [-]
Q	water uptake rate by a whole plant [mL min^{-1}]
θ	volumetric water content in soil [mL cm^{-3}]

θ_l soil water content by weight [mL g^{-1}]

r distance from root center [cm]

r_d distance at which initial radionuclide concentration is maintained [cm]

r_o root radius [cm]

R_f retardation factor [-]

R_{med} cancer risk avoided by restriction of cultivation [man]

R_{soc} crop yield lost by restriction of cultivation [kg-crop]

ρ soil particle density [g cm^{-3}]

ρ_b bulk soil density [g cm^{-3}]

s area [m^2]

S_c selectivity coefficient [-]

σ standard deviation [m]

t time [h or y]

T growth period [h]

T_c transpiration coefficient [mL g^{-1}]

T_f soil-to-plant transfer factor [Bq/g dry or wet vegetation per Bq g^{-1}-soil].

T_H half life of radioactive decay [y]

T_{life} life expectancy [y]

T_{Sr} half life of strontium-90(=28.8y)

v velocity of radial water movement [cm min^{-1}]

v_w water infiltration velocity through the soil [cm y^{-1}]

V_b water infiltration velocity per unit area without plants [L m^{-2} y^{-1}]

V_w water infiltration rate per unit area [L m^{-2} y^{-1}]

w water flux to xylem [mL h^{-1}]

w_e weight of the edible parts [g]

W total water uptake per plant [mL]

W_g weight of soil [kg]

W_l volume of solution [L]

W_{soil} areal surface soil density [kg cm^{-2}]

W_{water} volume of pure water added [mL]

X distance from the source on a horizontal scale [m]

Y distance from the source on a vertical scale [m]

Y_c annual crop yield per unit area [kg/m^2/y]

Y_{comp} compensation period [y]

Y_R restriction period [y]

CHAPTER 1. INTRODUCTION

1.1 BACKGROUND

Environmental engineers are often faced with the following question:

How clean is clean ?

The safety level of a toxic substance (pollutant) in the environment, which is referred to as an Environmental Quality Standard (EQS), has been determined on the basis of medical information such as the dose-response relationship to human health. The proper EQS cannot be established, however, without accurately tracing the pollutant transfer from the environment to humans. Usually, the pollutant behavior in an actual situation — transport in the atmosphere, deposition onto the soil surface, accumulation and migration in the soil, transfer to crops and animals, translocation to their edible parts, and transfer to humans — has been predicted with the help of mathematical models which are known as 'environmental transfer models'. If the respective environment has been specified properly and the information necessary for model prediction has been obtained, the established EQS will have a high reliability. In general, however, predictions are carried out without sufficient information on the complicated environmental factors affecting the pollutant behavior. Consequently, large uncertainties are associated with the calculated results. This leads to addition of a large margin for safety to each parameter so as not to underestimate the health risk. Although EQSs have already been set for many pollutants (Environmental Agency, 1992), the suitability of these values should be reexamined regularly with updated information on the behavior characteristics of each pollutant in the actual environment.

When the concentration is higher than the safety level, it becomes necessary to implement proper countermeasures such as cleanup, medical care for inhabitants, evacuation or relocation, control of food consumption, and restriction of agricultural activities (ICRP, 1984; IAEA, 1986b; IAEA, 1989a). In deciding which countermeasure should be selected, model precision is important because some countermeasures are accompanied by serious social and economic problems (IAEA, 1989a) which might have consquences more significant than the health risk. The derived intervention levels (DILs) should be as rational as possible; the rationality of the DIL needs to be discussed considering such social and economic

risks. Optimization of these complicated risks requires an accurate prediction of the long-term behavior of the pollutant, in other words, reliable environmental transfer models and reliable parameter values.

In the field of radioecology, accurate prediction of radionuclide transfer from soil to plants has been an important, but problematic subject. Difficulty in evaluating internal dose through a food chain was brought into focus by the Chernobyl accident. Large differences in the evaluated internal doses have been found between the data of the former USSR and those of the Advisory Committee though the external doses have been almost the same (IAC, 1991). This has been attributed to differences of the methods applied; the former USSR predicted the ingestion of ^{137}Cs using an environmental transfer model assuming no restriction on local food consumption, whereas the Committee used whole body counters and calculated the dose according to the metabolic information on the human body. Plots of the average whole body incorporation of ^{137}Cs in each inhabitant versus the concentration in the surface soil (**Figure 1**) have suggested that intake of ^{137}Cs by the residents was restricted almost in proportion to the concentration in the surface soil (International Advisory Committee, 1991). However, it seems doubtful that strict food control was carried out in accordance with the contamination level which had not been clarified before the measurement. It is possible that the tendency shown in the figure is attributable to the spatial difference of ^{137}Cs transfer characteristics from the soil to the agricultural products. Konshin (1992) has observed the inversely proportional relationship between the radiocesium concentration in the soil and the soil-to-plant transfer factor. A similar tendency has also been found in a soil-to-milk transfer (Knatko, et al., 1994). These results pose a question about the transfer factor concept which assumes linearity of concentration between environmental components. . More efforts to develop practical methods to predict precisely pollutant behavior in the environment are needed for both reliable risk evaluation and appropriate selection of countermeasures. In order to protect human health effectively from environmental pollution, it is of primary importance to improve the reliability of transfer models and parameter values used in environmental impact assessments.

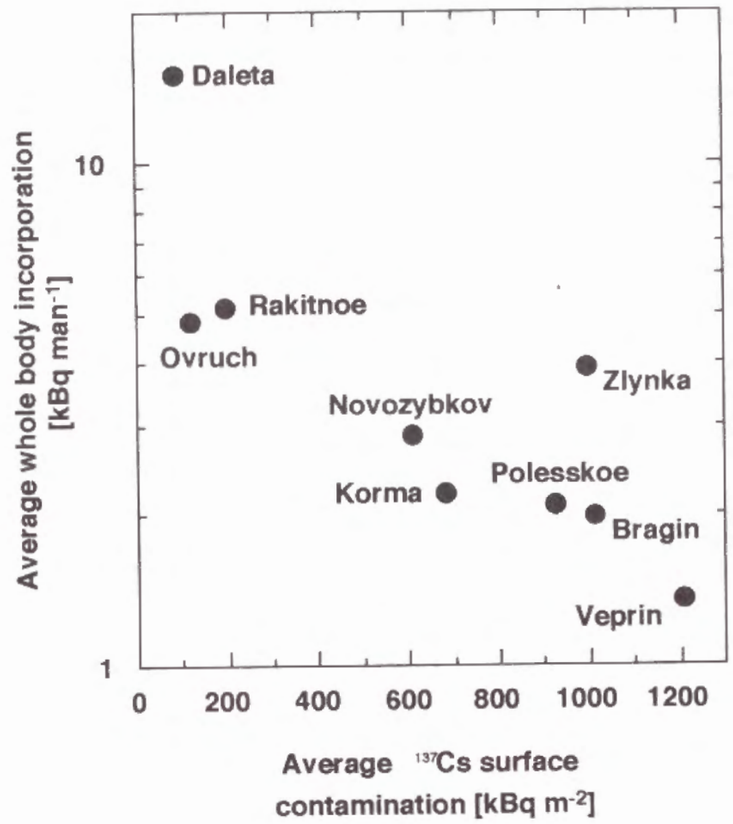

Figure 1. Relationship between measurements of cesium incorporated in the body of selected populations and official values for cesium surface contamination (from International Advisory Committee, 1991)

1.2 PURPOSE

The purpose of this study is reliability improvement of environmental transfer model in a soil-plant system. Although transfer models applied to this system have a very important role in evaluation of health risk due to intake of toxic substances released to the environment, the reliable prediction is still a problematic subject as mentioned above. In this system, there are many paths and environmental factors which vary greatly, affecting behavior of the pollutants (**Figure 2**). Uncertainties associated with environmental condition changes cannot be decreased without developing a practical method which can evaluate such variable effects. However, it does not seem that information on environmental conditions is being utilized for accuracy improvement of model prediction.

This dissertation discusses how to improve the reliability of selected model parameters by using qualitative and quantitative information on the environment. Two important parameters, soil-solution distribution coefficient (K_d) and soil-to-plant transfer factor (T_f), are focused on here because both have been obtained empirically and they are considered to have large uncertainties. Few studies have looked at the problem of reducing their uncertainties.

The objectives are summarized as follows.

- To review previous environmental transfer models in the soil-plant system for use in environmental impact assessments and clarify their features, similarities, and problems to be solved. (Chapter 2)
- To examine soil-solution distribution coefficients (K_ds), which are commonly used to evaluate the mobility of the radionuclides in soil, for their variation characteristics related to environmental conditions. (Chapter 3)
- To develop simple, practical methods to estimate radionuclide K_ds using the environmental information and to discuss the accuracy. (Chapter 3)
- To examine soil-to-plant transfer factors (T_fs) which are widely applied to prediction of radionuclide transfer through soil-crop pathway for their variation characteristics related to environmental conditions. (Chapter 4)
- To develop a new, practical model which can flexibly estimate T_f under changing environmental conditions and to discuss its accuracy. (Chapter 4)
- To present a decision-making process, with some new principles for

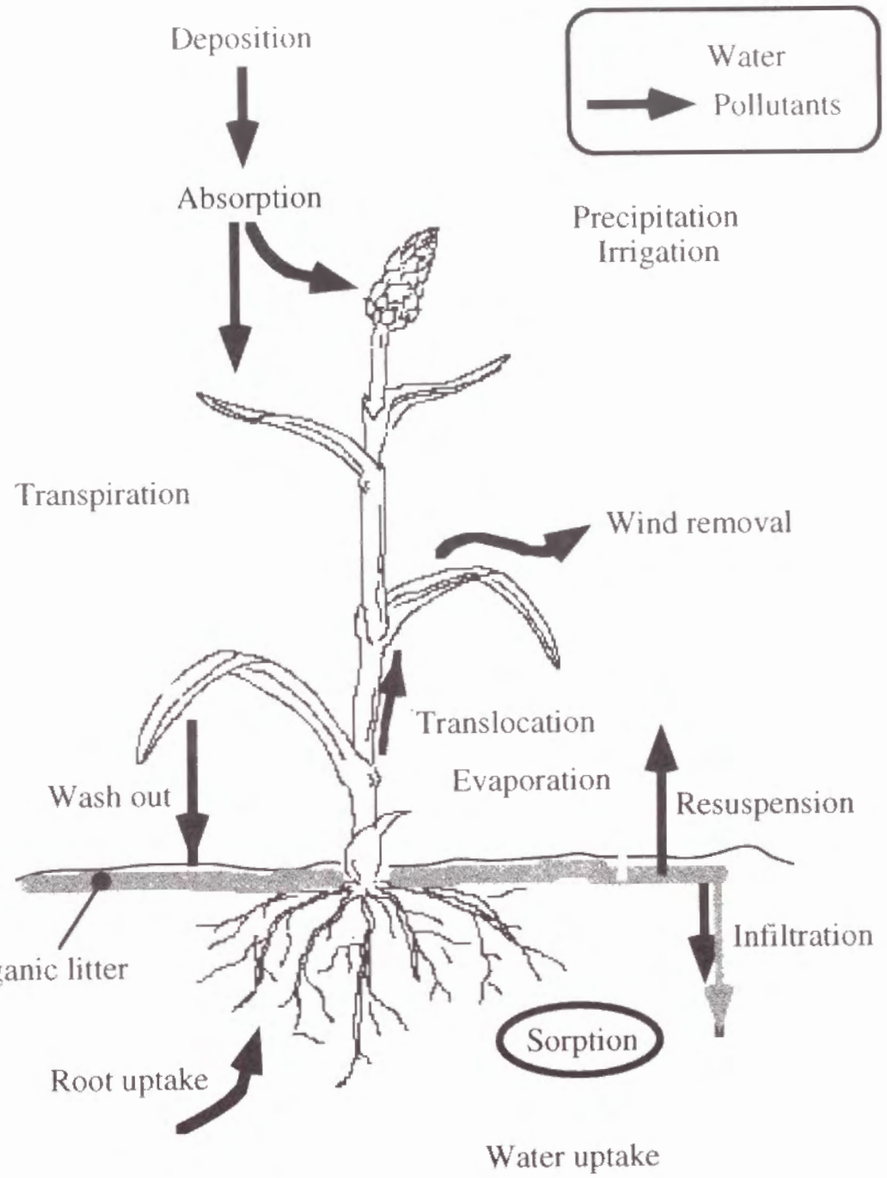

Figure 2. Pollutants and water migration in a soil-plant system; additionally, physical, chemical, and biological decays need to be considered.

intervention, with regard to soil contamination by utilizing the statistical information obtained above on K_ds and T_fs. (Chapter 5)

- To apply the methods and the model newly developed for the estimation of K_d and T_f to risk evaluation of soil contamination and to discuss their efficiency for reliability improvement in environmental impact assessments. (Chapter 5)

CHAPTER 2. ENVIRONMENTAL TRANSFER MODELS IN A SOIL-PLANT SYSTEM

2.1 INTRODUCTION

A terrestrial ecosystem is of primary importance in evaluating the risk of pollutants because we live in this ecosystem and most of our foods come from terrestrial sources. Short-term dilution as seen in an aquatic or atmospheric environment cannot be expected in this system because the soil, which is the main component, retains the pollutants for a long time. Additionally, physical, chemical, and biological reconcentration (bioaccumulation) can occur in the soil and, as a result, the pollutants become concentrated in some components, such as plants and animals, to much higher concentrations than initially.

When high amounts of pollutants have been released to the terrestrial ecosystem, the transfer to humans through a variety of pathways should be predicted in numerical ways. A mathematical model used for this purpose is referred to as an 'environmental transfer model'. This model enables us to evaluate the effects on human health and to supply necessary information for making decisions on which countermeasures should be carried out. However, it should be recognized that the calculation results obtained by an environmental transfer model have some uncertainty. Accurate prediction of pollutants in the terrestrial ecosystem cannot be achieved without sufficient information on the environmental conditions in real fields. In the following sections, the procedure for general model prediction is outlined and well-known environmental transfer models in a terrestrial ecosystem are briefly summarized with the values of the parameters involved.

2.2 PROCEDURE FOR MODEL PREDICTION

The model prediction process can be divided into five steps: scenario specification, model formulation, parameter selection, computerization (coding) and calculation, and documentation (**Figure 3**). This section explains the former three steps (scenario specification, model formulation, and parameter selection) which require careful consideration by environmental engineers.

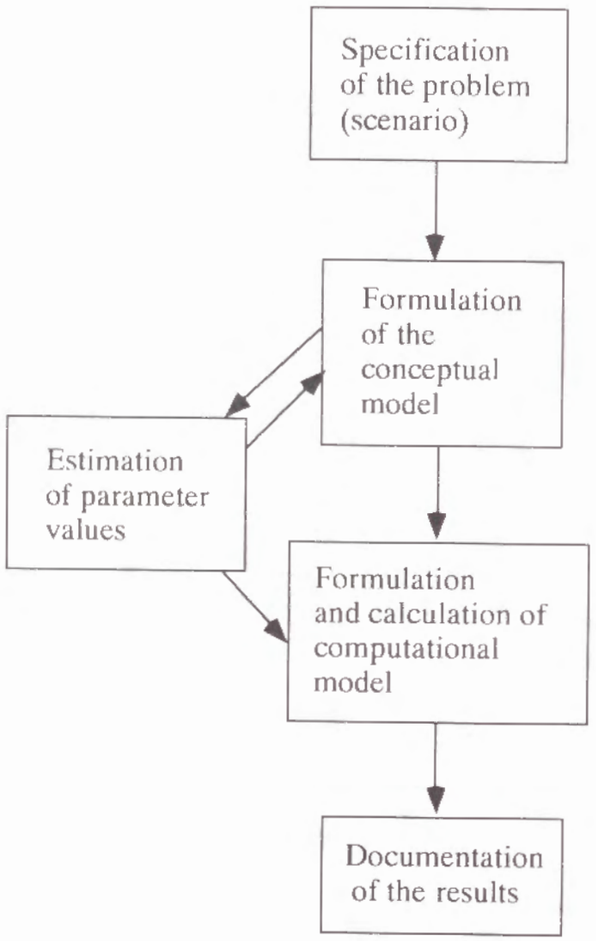

Figure 3. Diagram of model prediction process.

2.2.1 Scenario specification

The first step in risk evaluation is specification of the scenario under consideration. This means clarifying why, where, what, and for whom the evaluation should be done. The factors which must be considered are as follows:

a) The intended use of results (e.g. optimization of released amount / technological assessments / emergency planning)

b) The temporal resolution required (e.g. specification of the exposure period, consideration of the time of or after release, estimation of exposure dose rates or integrated exposure over the life time of the population)

c) The spatial resolution required (specification of site, critical population group, and regional variation of the identified population).

d) The degree of processing level (e.g. processes and mechanisms to be simulated).

e) The character of release (e.g. routine or planned releases / small or large scale accidents / future releases from a waste disposal site).

It is also desirable that social and economic factors be considered.

When cultivated lands become contaminated, the priority should be given to the integrated health risks to the consumers ingesting the agricultural products. In this case, reliability in air-crop (direct deposition) and soil-crop (root uptake) pathways become of primary importance. Selection of the parameters involved in these pathways, such as the removal rate constant from surface soil and the transfer factor from soil to crop, requires careful considerations.

2.2.2 Model formulation

According to the appropriately specified scenario, we formulate a conceptual environmental transfer model. For a peculiar environment such as an uniform soil layer, a precise model described by partial differential equations, like an advection-diffusion model, can be utilized. However, in a real assessment, such precise models are impractical because the times and areas under consideration are too large and we have insufficient information for short-term and microscopic model prediction. Then, in general assessments, the environment under consideration is represented by some major conceptual components called 'compartments'. The number of compartments and that of their interrelationships

change with the accuracy required. The quantities and qualities of available data also affect the model structure. The interrelationships among the compartments are described by differential equations. These equations are transformed to a computer code to simulate the pollutant behavior numerically. Parameter values and necessary data such as initial and boundary conditions need to be inputted before the calculation.

2.2.3 Parameter selection / estimation

Parameter values directly affect the calculated results. Even if the scenario is properly specified and the model is correctly formulated, a reliable prediction cannot be achieved without suitable parameter values. Unsuitable values arise from a lack of data, improper measurement techniques, and variations caused by environmental condition changes. Desirably these potential errors should be clarified through field tests under full range of conditions (Hoffman et al., 1984) in carefully designed experiments. However, that is a difficult and laborsome task.

Parameters included in environmental transfer models can be classified into the following four categories (IAEA, 1989b). Each term and examples of related parameters are as follows:

a) Source: starting time, duration, and rate of release, amount and composition of materials released, chemical and physical forms of each material.

b) Physical transport: wind speed and direction as a function of altitude, precipitation rate, temperature, water flow velocity based on hydro-geological features, and characteristics of material sorption onto soil.

c) Biological transfer: bioavailability of materials, transfer rate from soil or water to food products, agricultural practices, bioaccumulation in or decomposition by microorganisms, and loss from food products through weathering before harvest or processing after harvest.

d) Exposure: living and consumption habits, health status, age range and metabolic properties of population groups, and internal/external dose equivalents.

Many parameter values vary greatly with the conditions although they are usually presented without information on time, location, and other environmental factors. More efforts to collect information on the ambient conditions are necessary for the determination of proper, site-specific parameter values.

2.3 ENVIRONMENTAL TRANSFER MODELS

2.3.1 Outline

Many environmental transfer models have been developed in the field of radioecology to predict radiation doses to the human population. Some models have been widely used for use in radiological and environmental impact assessments required for regulatory compliance, facility design, and safety analyses. There are others which have been used for dose reconstruction of past fallout events. These models, however, are not the same in their structures; a large part of the difference is due to the situation to which each model is applied. The situations can be categorized into two types: normal operation or accidental release.

For safety assessment under normal operation, a long-term scale model, i.e., an 'equilibrium model' is generally used. This kind of model is called also 'concentration factor method' (ICRP, 1979) or 'steady-state model' (NCRP, 1984). The equilibrium model calculates the final, equilibrated concentrations of radionuclides in environmental components such as air, soil, crop, livestock, and humans. It consists of empirical, time-independent parameters and it has a good practicality because of its calculational simplicity. From the viewpoint of safety, conservative values have been generally selected for the parameters so as not to underestimate the actual effect.

A chronology showing development of these equilibrium models is given in **Figure 4**. The oldest equilibrium model referred to is HERMES (Soldat and Harr, 1971), which laid the foundation for many successors such as FOOD (Baker et al, 1976), USNRC Regulatory Guide 1.109 model (USNRC, 1977), Miller's model (Miller, et al., 1980), and IAEA Safety Series 57 model (IAEA, 1982). The USNRC model has been widely used and was succeeded by AIRDOS-EPA (Moore, et al., 1979) and Boone's model (Boone et al., 1981). United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR, 1972) proposed an empirically derived model for analyzing fallout data.

When radionuclides are accidentally released to the environment, rapid prediction of the transfer and accumulation of the radionuclides in the environment and reliable evaluation of the possible maximum dose to the the

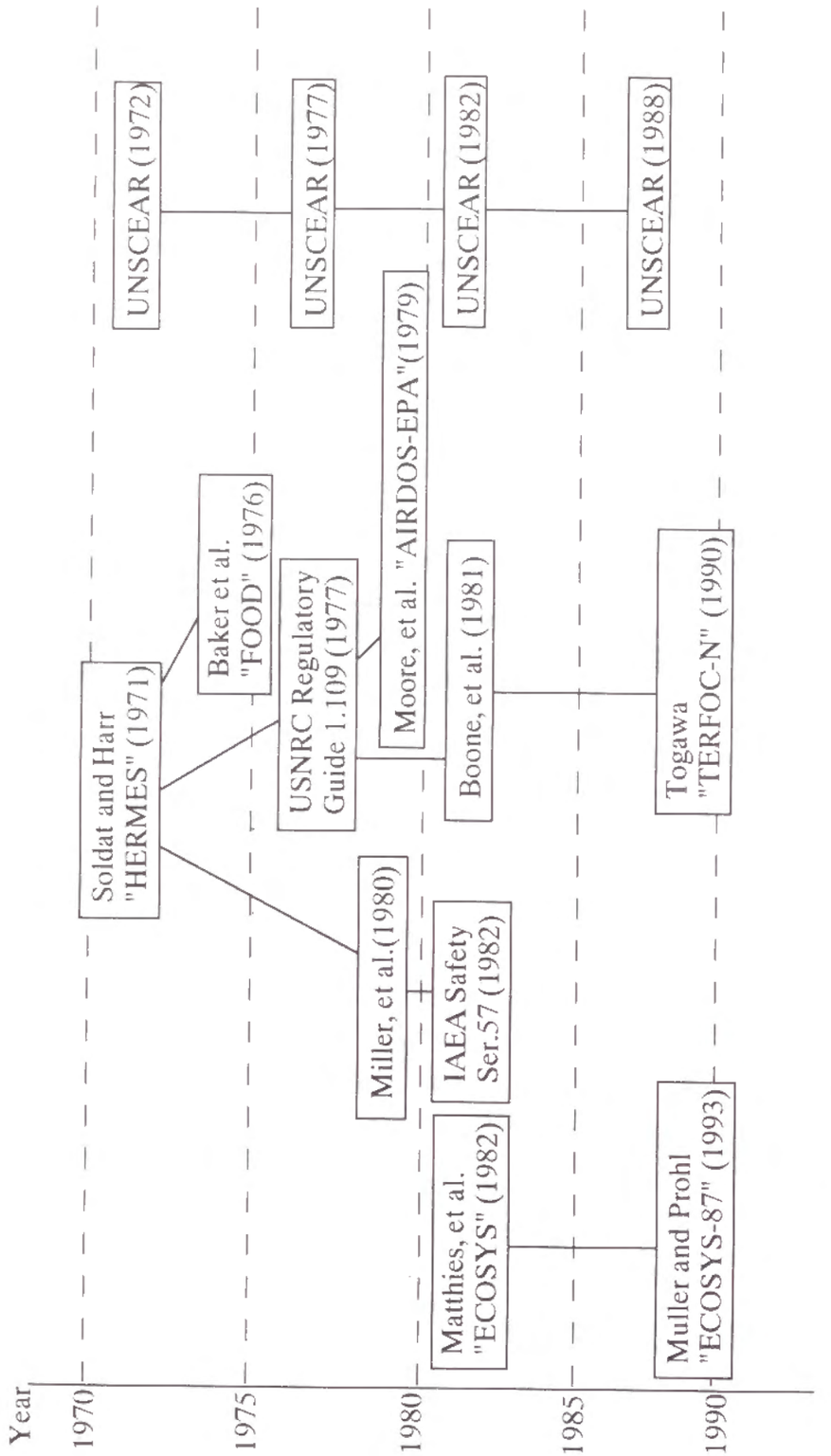

Figure 4. Chronology showing development of selected equilibrium models.

public are of primary importance. To such an emergency situation, a short-term scale model, i.e., 'dynamic model' needs to be applied. This kind of model is called also as 'systems analysis method' (ICRP, 1979) or 'transient model' (NCRP, 1984). In this dynamic model, radionuclide transfers between environmental components are described by transfer rates and the concentration in each component is given as a function of time. Although its structure and calculation procedure are complicated compared to those of the equilibrium model, the dynamic model provides more flexibility in considering the chronological changes of the environmental and growth conditions, which leads to more realistic results.

A chronology showing development of these dynamic models is given in **Figure 5**. The oldest dynamic model referred to is TERMOD (Booth and Kaye, 1971), which has been followed by many models such as WASH-1400 model (USNRC, 1975), NRPB-R89 model (Simmonds, et al., 1979), RADFOOD (Koch and Tadmor, 1986), PATHWAY (Whicker and Kirchner, 1987), and COMIDA (Abbott and Rood, 1993).

In the following, selected transfer models are briefly described focusing on air-crop-soil pathway. They appear in chronological order for each category. The symbols used here may not be those originally used; changes were made to assist the reader in understanding their meanings and for consistency. The models for gaseous nuclides (e.g. tritium) and carbon are not considered.

2.3.2 Equilibrium models.

HERMES (1971). Although some transfer models appeared in the1960's (e.g. Ng and Thompson, 1966), HERMES (Soldat and Harr, 1971) has been widely accepted as the first of the equilibrium models. HERMES was developed for a study on potential radiation doses to people from nuclear facilities situated in the upper Mississippi River basin in the year 2000. In HERMES, radionuclides were assumed to reach food crops through direct deposition from air and sprinkler irrigation, and uptake from soil (**Figure 6**). The equation to calculate radionuclide concentration in a crop was as follows:

$$C_{crop} = \frac{f \cdot f_{ret}}{Y_c} \cdot \sum_{m=a}^{h} \left[\left(Q_{c\_dep} + r_{irr} \cdot C_{irr} \right) \cdot e^{-\lambda \cdot t} \right] + \frac{T_f}{W_{soil}} \cdot \left(Q_{s\_air} + Q_{s\_irr} \right) \qquad (1)$$

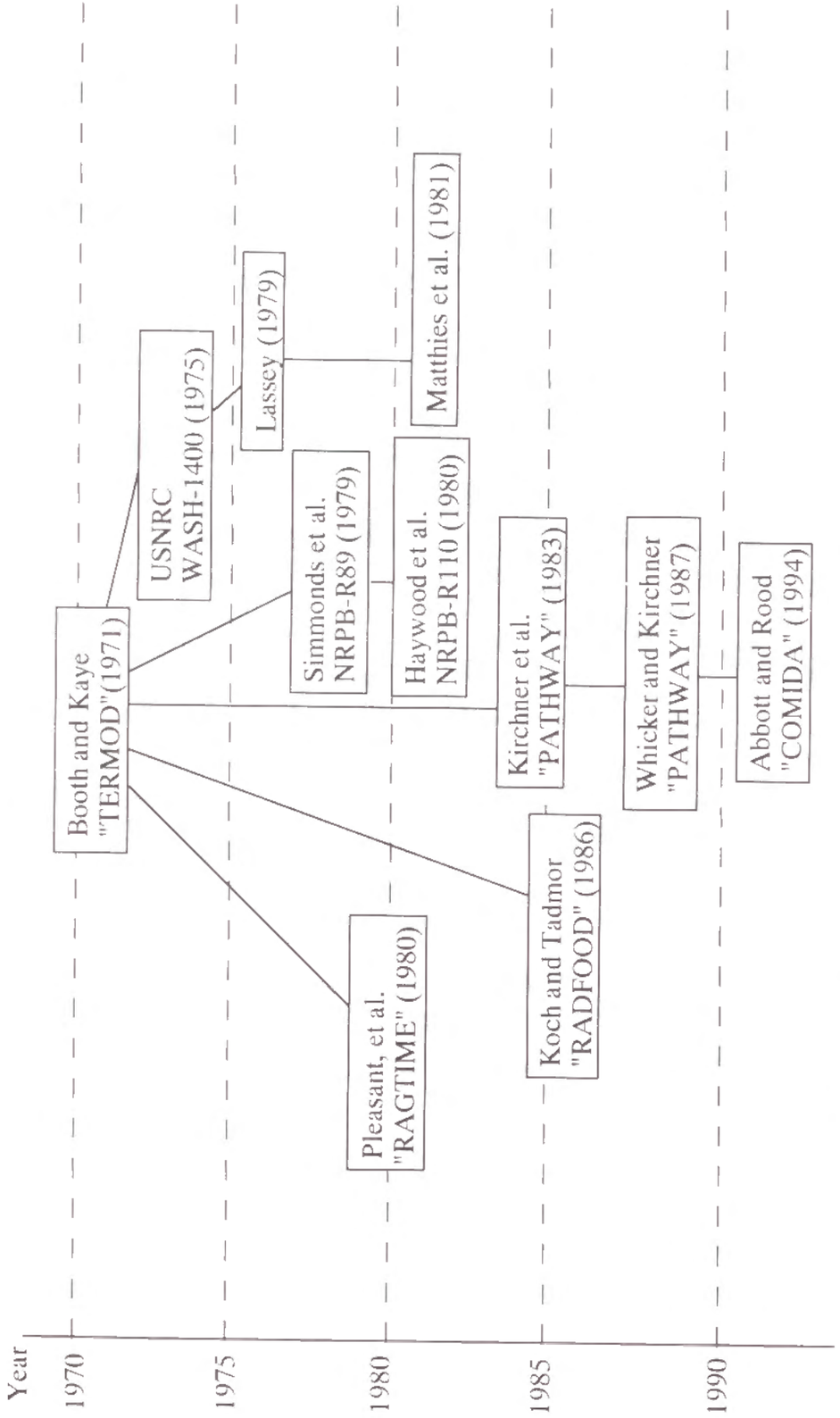

Figure 5. Chronology showing development of selected dynamic models.

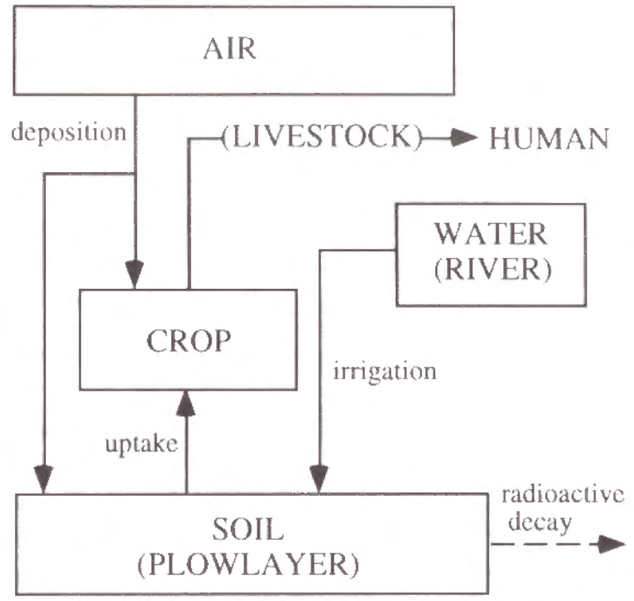

Figure 6. Mass flow diagram of HERMES in soil-plant system.

where f is the translocation factor to edible parts [-]; f_{ret}, the fraction of deposited activity retained on crops (the deposition retention factor) [-]; Y_c, the crop yield [kg m^{-2}]; a, the month of appearance; h, the month of harvest; Q_{c_dep}, the deposition amount on crop per unit soil surface area [Bq m^{-2}]; r_{irr}, irrigation rate during month 'm' [L m^{-2}]; C_{irr}, concentration in the irrigation water during month 'm' [Bq L^{-1}]; λ, the effective decay constant [d^{-1}]; t, the time between deposition and harvest [d]; T_f, the transfer factor from soil to crop edible parts at time of harvest [Bq kg^{-1}-crop per Bq kg^{-1}-soil]; W_{soil}, the effective density of surface soil (plowlayer) [kg m^2]; Q_{s_air}, the concentration in plowlayer of the radionuclide deposited from air [Bq m^{-2}]; Q_{s_irr}, the concentration in plowlayer of the radionuclide deposited from water (irrigation) [Bq m^{-2}].

HERMES was referred to in successor food chain models such as FOOD (Baker et al, 1976), USNRC Regulatory Guide 1.109 model (USNRC, 1977), Miller's model (Miller, et al., 1980), and IAEA Safety Series 57 model (IAEA, 1982). Among them, the USNRC model has been widely accepted to radiological assessments.

USNRC Regulatory Guide 1.109 model (1977). The USNRC Regulatory Guide 1.109 model (USNRC, 1977) followed the basic structure of HERMES. USNRC model was intended to estimate maximum radiation doses to the population within 50 miles around a light-water-cooled nuclear power plant (LWR). The equation for predicting radionuclide intake by humans through irrigated crop was described as

$$I_{crop} = U_{crop} \cdot d \cdot \exp(-\lambda_d \cdot t_{hold}) \cdot f_{DE} \cdot \left[\frac{f_{ret} \cdot \left(1 - \exp\left(-\lambda_{crop} \cdot t_{cexp}\right)\right)}{Y_c \cdot \lambda_{crop}} + \frac{f_{irr} \cdot T_f \left(1 - \exp\left(-\lambda_d \cdot t_{sexp}\right)\right)}{W_{soil} \cdot \lambda_d} \right] \quad (2)$$

where I_{crop} is the radionuclide intake by an individual [Bq]; U_{crop}, the usage (consumption) factor that specifies the exposure time or intake rate for an individual [kg y^{-1}]; d, the radionuclide deposition rate [Bq m^{-2} h^{-1}]; λ_d, the radioactive decay constant [h^{-1}]; t_{hold}, the holdup time that represents the time interval between harvest and consumption of the food [h]; f_{DE}, the dose conversion factor, which can be used to calculate the radiation dose from intake of radionuclides [Sv Bq^{-1}]; f_{ret}, the fraction of deposited activity retained on crops [-]; λ_{crop}, the effective removal rate constant from crops [h^{-1}] ($=\lambda_d+\lambda_{we}$); λ_{we}, the removal rate constant of physical loss by weathering [h^{-1}]; t_{cexp}, the time period that crops are exposed to contamination during the growing season [hr]; Y_c, the agricultural productivity (crop yield) [kg m^{-2}]; f_{irr}, the fraction of the year the crops are irrigated [-]; T_f, the transfer factor from soil to crop edible parts at time of harvest [Bq kg^{-1}-crop per Bq kg^{-1}-soil]; t_{sexp}, the time period for which soil is exposed to contamination [h]; W_{soil}, the effective surface soil density [kg m^{-2}].

In USNRC model, the radionuclide concentration in soil decreased only by radioactive decay; downward movement to subsurface soil was not considered.

UNSCEAR model (1972). Many fallout data on the radionuclides emanating from nuclear weapon tests had been collected worldwide. The United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR, 1972) analyzed these data using the empirical equation:

$$M_n = P_1 F_n + P_2 F_{n-1} + P_3 \sum_{m=1}^{x} F_{n-m} \cdot e^{-\mu \cdot m} \quad (3)$$

where M_n is the predicted concentration of the radionuclide in a particular food

stuff, averaged during the nth year [Bq kg^{-1}]; F_n, the total annual fallout during the nth year [Bq m^{-2}]; the P_1, P_2, P_3, and μ are constants giving the best fits to the relationship. The terms P_1 and P_2 are usually interpreted as representing short-term pathway to foodstuffs like direct deposition onto foliar surfaces of crops.

The results predicted by eq.3, however, have large uncertainty due to condition changes. An alternative model to the UNSCEAR model is that of Bartlett et al. (1972).

Boone's model (1981). Boone et al. (1981) extended the USNRC model using the data collected for specific USA southern crops and livestock (Ng et al., 1978; 1979a; 1979b). The mass flow diagram of the Boone's model is shown in **Figure 7**. In this model, the radionuclide concentration in crop edible parts was calculated by

$$C_{crop} = \frac{f \cdot d}{Y_c} \cdot \int_0^{t_{exp}} \left[1 - e^{-M (Y_c / t_{cexp}) t}\right] \cdot e^{-\lambda_{crop} \cdot (t_{cexp} - t)} \, dt + \left[1 - f_g + f_g \cdot f_{soil}\right] \cdot \frac{d \cdot T_f}{W_{soil} \cdot \lambda} \cdot \left(1 - e^{-\lambda \cdot t_b}\right) \quad (4)$$

where C_{crop} is the concentration in crop edible parts [Bq kg^{-1}]; f, the translocation factor, i,e. the concentration ratio of radionuclide in the edible parts to that in the total above-ground parts of the crop at the time of harvest [-]; d, the deposition rate [Bq m^{-2} $month^{-1}$]; Y_c, the total, recoverable above-ground crop yield [kg m^{-2}]; t_{cexp}, the growing or exposure period [month]; λ_{crop}, the effective removal rate from crops [$month^{-1}$] ($=\lambda_d+\lambda_{we}$); λ_d, the radioactive decay constant [$month^{-1}$]; λ_{we}, the removal rate due to weathering [$month^{-1}$]; f_g, the fraction of year during which the crops are grown [-]; f_{soil}, the fraction of deposited activity transferred to soil [-]; T_f, the transfer factor from soil to crop edible parts at time of harvest [Bq kg^{-1}-crop per Bq kg^{-1}-soil]; W_{soil}, the effective surface soil density [kg m^{-2}]; λ, the effective removal rate constant [$month^{-1}$]($=\lambda_d+\lambda_i+\lambda_c$); λ_i, the downward migration rate into the region below the root zone [$month^{-1}$]; λ_c, the root uptake rate by the crops [$month^{-1}$]; t_b, the time of long-term buildup [month]; M, the proportionality constant [-] which is related to the fraction of initial deposition retained on the crop (f_{ret}) as follows (Chamberlain, 1970):

$$f_{ret} = 1 - e^{-M \cdot Y_c} \quad (5)$$

Based on the Boone's model, TERFOC-N (Togawa et al., 1990) was developed for application to not only LWRs but also other nuclear facilities.

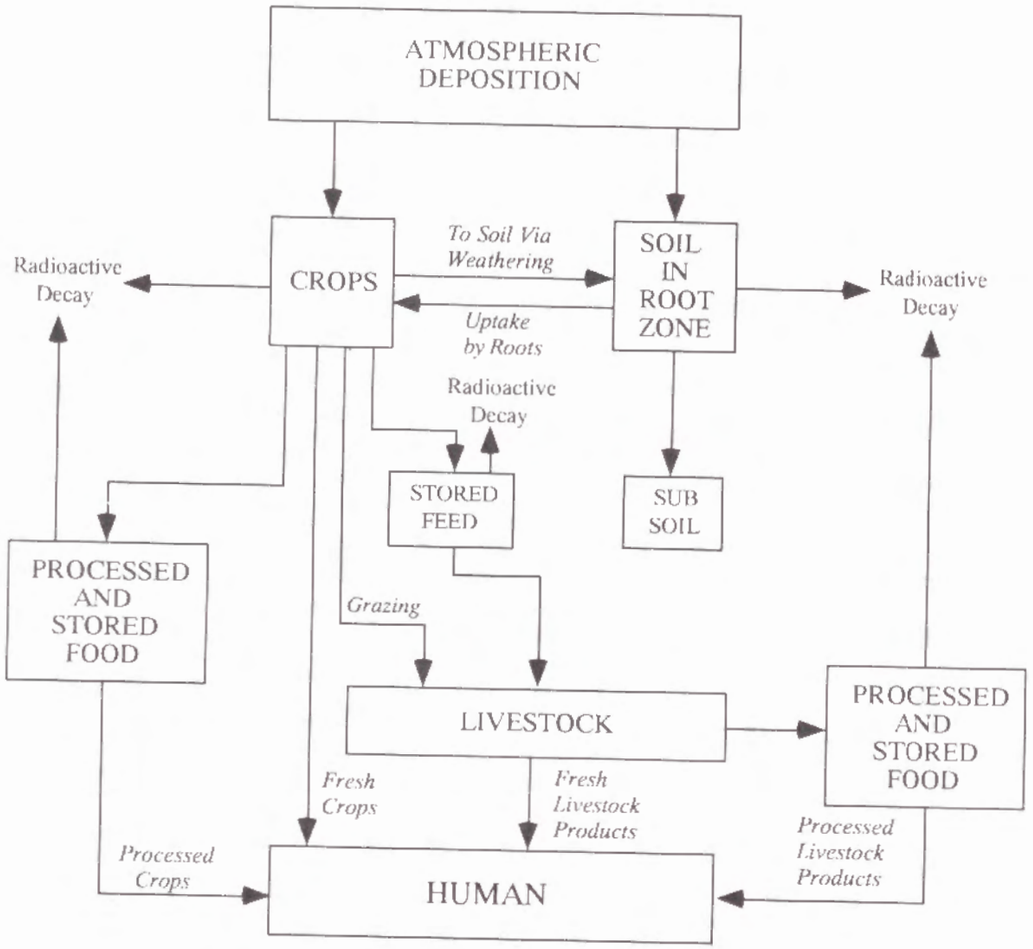

Figure 7. Mass flow diagram of Boone's model.

ECOSYS-87 (1993). ECOSYS-87 (Muller and Prohl, 1993), an improved version of ECOSYS (Matthies et al., 1982), has the structure of an equilibrium model although it was referred to by its authors as a 'dynamic model'. ECOSYS-87 was developed after the Chernobyl accident which had demonstrated the need for more differentiation in food chain modeling. ECOSYS-87 considered 18 plant species, 11 animal products, and 18 processed products. Exposure doses through ingestion and inhalation were estimated for six age groups (**Figure 8**).

In ECOSYS-87, the concentration of radioactivity in a plant harvested at time=t after deposition ($C_{crop}(t)$ [Bq kg^{-1}]) was given by

$$C_{crop}(t) = \frac{d_{crop}}{Y_c} \cdot f(t) \cdot \exp\left[-(\lambda_{we} + \lambda_d) \cdot t\right] + T_f \cdot C_{soil}(t) \qquad (6)$$

where d_{crop} is the total deposition onto the crop [Bq m^{-2}]; Y_c, the crop yield at the harvest [kg m^{-2}]; f(t), the translocation factor given as a function of time [-]; λ_{we}, the weathering loss rate due to rain and wind [d^{-1}]; λ_d, the radioactive decay constant [d^{-1}]. T_f, the soil-to-plant transfer factor [Bq kg-crop^{-1} per Bq kg-soil^{-1}]; $C_{soil}(t)$, the concentration of radioactivity in the soil. The f(t) was assumed to be 1 for the plants entirely consumed. The λ_{we} was assumed to be zero for those partly consumed. The C_{soil} was given by

$$C_{soil}(t) = \frac{d_{soil}}{d_s \cdot \rho_b} \cdot \exp\left[-(\lambda_i + \lambda_{fix} + \lambda_d) \cdot t\right] \qquad (7)$$

where d_{soil} is the total deposition to soil [Bq m^{-2}]; d_s, the depth of root zone [m]; ρ_b, the bulk soil density [kg m^{-3}]; λ_i, the migration rate out of the root zone [day^{-1}]; λ_{fix}, the fixation rate in soil [day^{-1}].

ECOSYS-87 has been available since 1987 and has been utilized in emergency management systems in Germany (Jacob et al., 1991). This model has been utilized to recent studies on model validation such as BIOMOVS (Haegg and Johansson, 1988; Kohler, et al., 1991) and VAMP (Linsley, et al., 1990).

2.3.3 Dynamic models

In the dynamic models, the kinetics of the radionuclide transfer is described by a set of linear first-order differential equations as follows:

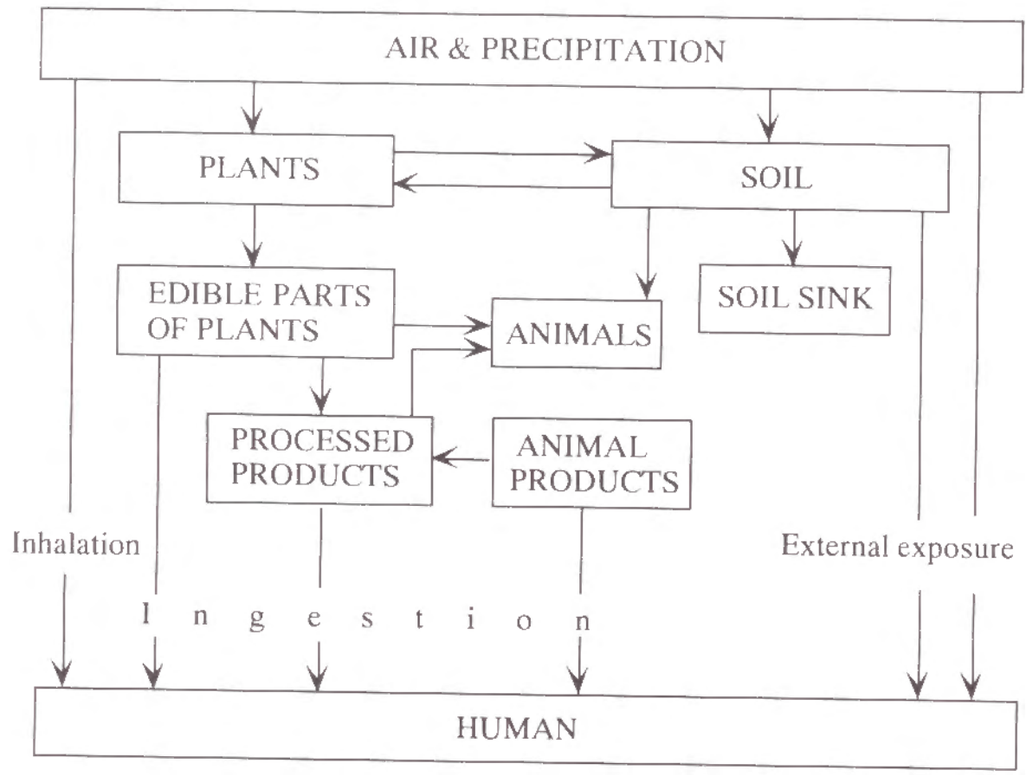

Figure 8. Mass flow diagram of ECOSYS-87.

$$\frac{dQ^k}{dt} = \sum_j^n \lambda_r^{jk} \cdot Q^j - \left\{ \sum_j^n \lambda_r^{kj} + \lambda_d \right\} \cdot Q^k + S_k \qquad (8)$$

where Q^k is the quantity of radionuclide in the component k [Bq kg^{-1}]; λ^{jk}, the transfer rate from component-j to component-k [T^{-1}]; λ_d, the radioactive decay constant [T^{-1}]; and S_k, the formation (generation/sink) constant [Bq kg^{-1} T^{-1}]. The relationships among the environmental components, however, are described differently for each model.

TERMOD (1971). TERMOD (Booth and Kaye, 1971) is known as the first model using a systems analysis technique. In this model, food crops and grass were dealt with separately; feedback from soil to plant was considered for only grass (**Figure 9**).

TERMOD described the change of radionuclide amount in food crops as follows:

$$\frac{dQ_{crop}}{dt} = f_{ret} \cdot d - \left(\lambda_d + \frac{\lambda_{cm}}{A_{cm}} + \lambda_{cs} \right) \cdot Q_{crop} \qquad (9)$$

where Q_{crop} is the radioactivity present on above-surface parts of the crop per square meter of the ground surface on which the crop is grown [Bq m^{-2} T^{-1}]; f_{ret}, the fallout correction factor to account for different depositions to the above-surface food [-]; d, the fallout source, which was assumed to be present only at time =0 [Bq m^{-2} T^{-1}]; λ_d, the radioactive decay constant [T^{-1}]; λ_{cm}, the transfer coefficient from food crops to man [m^2 T^{-1}]; A_{cm}, the soil surface area required to furnish food crops per one man [10^3 m^2]; and λ_{cs}, the transfer coefficient from the surface of above-ground parts to the soil surface [T^{-1}].

For grass, the feedback from soil to grass was added to the previous model as follows:

$$\frac{dQ_{grass}}{dt} = f_{ret} \cdot d - \left(\lambda_d + \lambda_{gs} + \frac{M_{cow}}{A_{grass} \cdot W_{grass}} \right) \cdot Q_{grass} + \lambda_{sg} \cdot Q_{soil} \qquad (10)$$

where Q_{grass} is the radioactivity present in the grass compartment [Bq m^2]; λ_{gs}, the transfer coefficient from grass to soil [T^{-1}]; M_{cow}, the rate of grass consumption by a cow [kg-dry T^{-1}]; A_{grass}, the pasture area per cow [m^2]; W_{grass}, the amount of grass

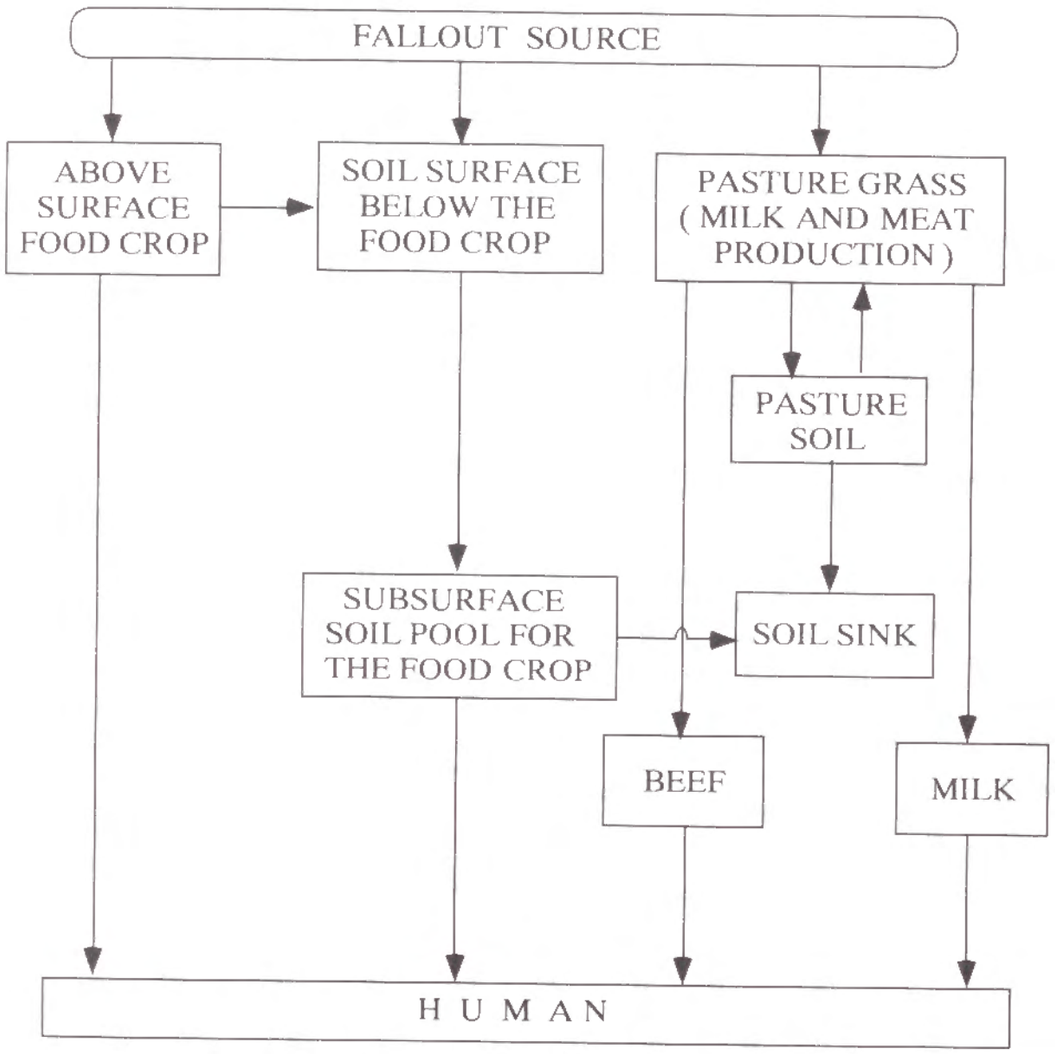

Figure 9. Mass flow diagram of TERMOD in a soil-plant system.

per unit area [kg m^{-2}]; λ_{sg}, the transfer coefficient from soil to grass [T^{-1}]; and Q_{soil}, the radioactivity present in the soil from the ground surface to the root depth of the grass [Bq m^{-2}]. In calculation, conservative values (Ng et al.,1968) were incorporated from the viewpoint of safety.

TERMOD laid the basis for many successor models such as the WASH-1400 model (USNRC, 1975), NRPB R-89 model (Simmonds, et al., 1979), RAGTIME (Pleasant, et al., 1980), RADFOOD (Koch and Tadmor, 1986), and PATHWAY (Whicker and Kirchner, 1987).

NRPB R-89 model (1979).

In the NRPB report 89, Simmonds et al.(1979) presented a dynamic model in which soil and plant compartments were divided into smaller parts. The soil was assumed to be well mixed for food crops and undisturbed for pasture grass. The undisturbed soil was divided into several compartments by depth. Resuspension and inadvertent soil consumption by livestock was assumed to occur only from the surface (uppermost soil) compartment. The plant was divided into two parts: external and internal plants. The transfer to the internal plant was expressed by two pathways: root-uptake and translocation from external surfaces on which radionuclides had deposited.

Haywood et al.(1980) revised the NRPB R-89 model using the fallout data collected in the U.K. to take into account the observed process of fixation of cesium in soil and plant-base absorption of strontium. Simmonds and Linsley (1981) validated Haywood's model using data on ^{90}Sr concentrations in milk collected in the U.K.

A similar, but original dynamic model was also proposed by Thorne and Coughtrey (1983). They divided the soil into ten layers with each 3 cm thick consisting of three components: soil solution, organic matter, and inorganic matter. The plants were separated into six parts: root, root-store, stem, internal leaf, external leaf, and grain or fruit.

RADFOOD (1986). The RADFOOD (Koch and Tadmor,1986) shown in **Figure 10**, which was developed on the basis of TERMOD (Booth and Kaye, 1971), considered six crop groups: forage crops, underground vegetables, leafy

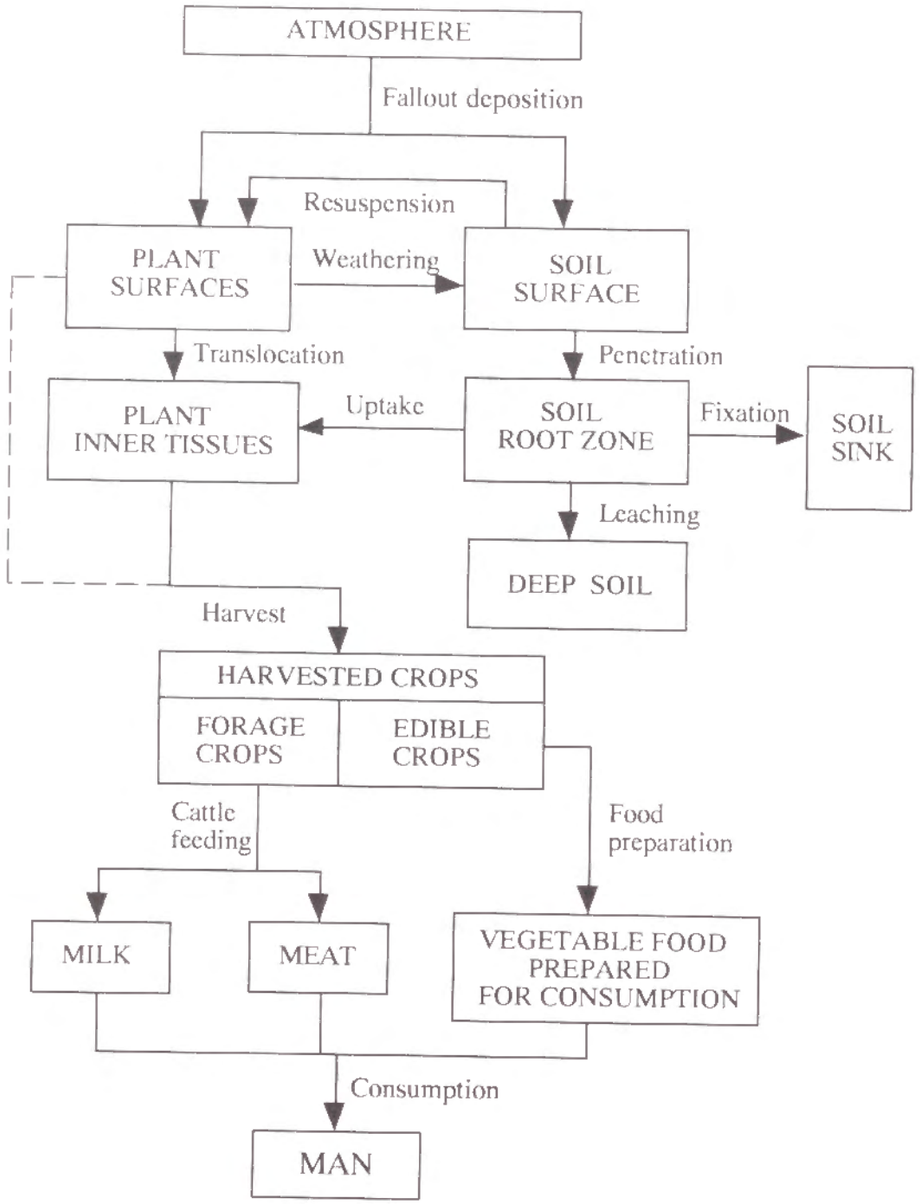

Figure 10. Mass flow diagram of RADFOOD.

vegetables, fruit-bearing vegetables, fruits, and cereals. The radionuclide concentration in the edible inner parts of each crop (C_{crop} [Bq kg^{-1}]) was described by

$$\frac{dC_{crop}}{dt} = -\lambda_d \cdot C_{crop} + \frac{\lambda_{up}}{Y_c} \cdot Q_{soil} + \frac{\lambda_f \cdot Y_{ex}}{Y_c} \cdot C_{leaf} \qquad (11)$$

where λ_d is the radioactive decay constant [d^{-1}]; λ_{up}, the rate constant of root uptake [d^{-1}]; Y_c, the yield of the edible parts [kg m^{-2}]; Q_{soil}, the radioactivity in the root-zone soil [Bq m^{-2}]; λ_f, the rate constant of translocation to edible parts [d^{-1}]; Y_{ex}, the yield of the external (non-edible) parts [kg m^{-2}]. The Q_{soil} and C_{leaf} were represented by the following equations:

$$\frac{dQ_{soil}}{dt} = -(\lambda_i + \lambda_{up} + \lambda_d) \cdot Q_{soil} + \lambda_{ss} \cdot Q_{surface} \qquad (12)$$

$$\frac{dC_{leaf}}{dt} = -(\lambda_{we} + \lambda_f + \lambda_d) \cdot C_{leaf} + \frac{\lambda_{res} \cdot f_{ret}}{Y_{ex}} \cdot Q_{surface} \qquad (13)$$

where λ_i is the rate constant of transfer to soil sink [d^{-1}]; λ_{ss}, the rate constant of percolation into root-zone soil (Q_{soil}) from the soil surface [d^{-1}]; $Q_{surface}$, the radioactivity in the soil surface layer [Bq m^{-2}]; λ_{we} is the weathering rate constant from the leaves [d^{-1}]; λ_{res}, the resuspension-deposition rate constant [d^{-1}]; f_{ret}, the fallout interception factor of leaves [-]. The initial C_{leaf} (C_{leaf_0}) was given by

$$C_{leaf\_0} = \frac{f_{ret} \cdot F_0}{Y_{ex}} \qquad (14)$$

where F_0 is the initial fallout radioactivity per unit soil surface [Bq m^{-2}].

PATHWAY (1983; 1987). The complete version of PATHWAY was described by Whicker and Kirchner (1987) following an incomplete description 4 years earlier (Kirchner et al., 1983). This agricultural food-chain model was applied to reexamination of radiation doses to residents in the region surrounding the Nevada Test Sites (NTS).

In the soil-plant system, PATHWAY set five compartments: vegetation surface (radioactivity: Q_{vs} [Bq m^{-2}]), vegetation internal tissues (Q_{vi} [Bq m^{-2}]), surface soil (Q_{ss} [Bq m^{-2}]), labile (active root-zone) soil (Q_{rs} [Bq m^{-2}]), and fixed soil (Q_{fs} [Bq m^{-2}]) (**Figure 11**). The radioactivities in these compartments were calculated by the following equations:

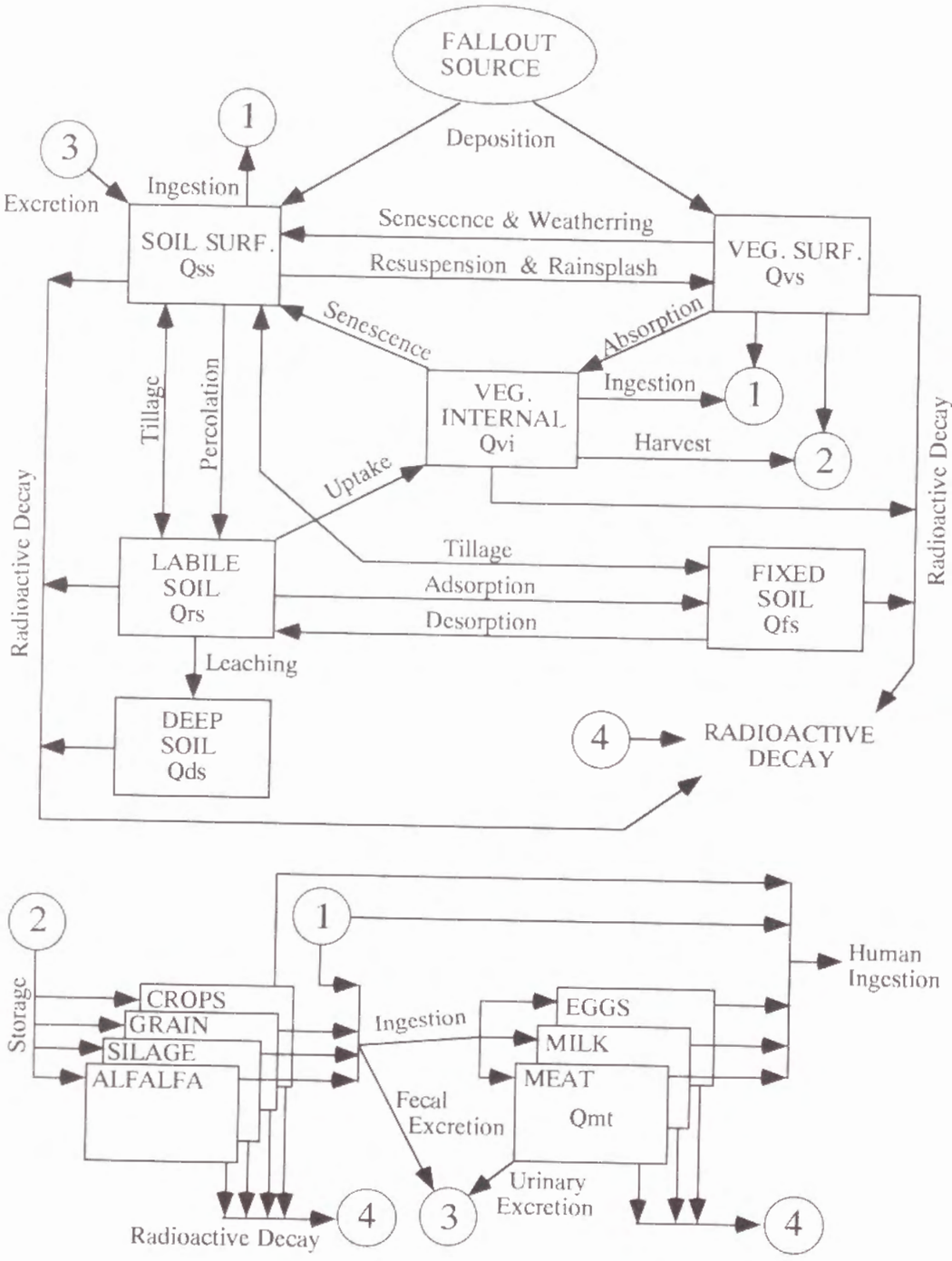

Figure 11. Mass flow diagram of PATHWAY; circled numbers connect processes between the upper and lower diagrams.

$$\frac{dQ_{vs}}{dt} = (\lambda_{res} + \lambda_{spl}) \cdot Q_{ss} - (\lambda_{we} + \lambda_{d} + \lambda_{f}) \cdot Q_{vs} \qquad (15)$$

$$\frac{dQ_{vi}}{dt} = \lambda_{f} \cdot Q_{vs} + R_{up} - (\lambda_{d} + \lambda_{sen}) \cdot Q_{vi} \qquad (16)$$

$$\frac{dQ_{ss}}{dt} = \lambda_{we} \cdot Q_{vs} - (\lambda_{ves} + \lambda_{spl} + \lambda_{per} + \lambda_{d}) \cdot Q_{ss} \qquad (17)$$

$$\frac{dQ_{rs}}{dt} = \lambda_{per} \cdot Q_{ss} + \lambda_{des} \cdot Q_{fs} - (\lambda_{i} + \lambda_{fix} + \lambda_{d}) \cdot Q_{rs} - R_{up} \qquad (18)$$

$$\frac{dQ_{fs}}{dt} = \lambda_{fix} \cdot Q_{rs} - (\lambda_{des} + \lambda_{d}) \cdot Q_{fs} \qquad (19)$$

where λ_{res} is the resuspension rate constant [d^{-1}]; λ_{spl}, the rainsplash rate constant [d^{-1}]; λ_{we}, the weathering rate constant [d^{-1}]; λ_{d}, the decay rate constant [d^{-1}]; λ_{f}, the foliar absorption (translocation from the surface to the internal part) rate constant [d^{-1}]; R_{up} is the root uptake rate [Bq m^{-2} d^{-1}]; λ_{sen}, the senescence rate constant [d^{-1}]; λ_{per} is the percolation rate constant [d^{-1}]; λ_{i} is the leaching rate constant [d^{-1}]; λ_{fix}, the fixed soil adsorption rate constant [d^{-1}]; λ_{des}, the fixed soil desorption rate constant [d^{-1}].

PATHWAY was validated using the data collected in the Smallboy detonation at the NTS and it gave the encouraging results for predinction of ^{131}I concentration in milk (Whicker et al., 1990). In comparison with selected previous models using Chernobyl data, however, PATHWAY underpredicted the grass/air and milk/air concentration ratios (Ng and Hoffman, 1988).

COMIDA (Abbott and Rood, 1994), having almost the same structure as that of PATHWAY, was designed to support the new USDOE version of the MELCOR Accident Consequence Code System (MACCS) (Jow et al. 1990).

2.3.4 Problems and future needs

All the models are imperfect. We need to keep in mind that both equilibrium and dynamic models have limitations in accuracy. All models are accompanied by some uncertainties due to lack of information and potential variabilities (IAEA, 1989b). A number of errors may possibly occur in the model prediction process: scenario specification, model formulation, parameter selection, computerization (coding) and calculation, and documentation (Figure 3). Even if the numerical

errors in computerization and calculation are avoided by careful verification, more considerations are necessary in order to reduce the other errors.

In scenario specification, the factors listed in section 2.2.1 must be given deterministically. An improper scenario can result in a correct answer for the wrong question. The perfect scenario, however, is difficult to establish because of lacking information, such as missing pathways, global-scale changes of environmental conditions, and unexpected events. It is desired to reduce these errors by collecting necessary information such as nuclide behavior characteristics, long-term condition changes, and probabilities of possible events.

In the model formulation, errors due to simplification (formulation of a compartment-type model) cannot be avoided and undetected flaws may be present. These models might have undetected flaws. The existing models, however, have not been tested sufficiently to validate their structures. The parameters involved might not be independent of each other, but co-variable. Such interrelationships should be categorized as 'structure errors' and need to be corrected by reformation of the model. For example, an inverse correlation has been found between K_d and T_f (Baes, 1982; Peterson, 1983; Baes et al., 1984; Sheppard and Evenden, 1988). Yasuda and Inoue (1992) described the soil-to-plant transfer factor (T_f) using the distribution coefficient (K_d) and the selectivity coefficient (Dorp et al., 1979). Transfer rate of nuclides from soil solution to plants was found to be non-linearly related to the concentration in solution (Shaw et al., 1992); this fact does not agree with the transfer factor concept which is based on the linearity of the concentration between the two compartments. An apparent non-linear relationship between soil-to-plant transfer factors and the concentration in soil has also been observed by Konshin (1992).

Errors might also be made in the process of transforming differential equations to computer codes. To find and eliminate the bugs (mistakes in coding), the code should be tested and verified many times; it is effective to compare the calculation results with analytical solutions.

In the parameter selection, we need to keep in mind that any environmental parameter has a variability to some extent due to regular or irregular changes of environmental conditions. How to deal with such variability has been a problematic subject. So as not to underestimate the risk of exposure to humans

(for safety analysis), sufficient margins need to be added to these variable parameters. Suitability of these margins, however, needs to be discussed on the basis of variational characteristics of each parameter. It seems inevitable that we depend on a probabilistic method if it is applicable. Statistical information on the parameters should be effectively applied to evaluation of the potential variability of the predicted results. Matthies et al. (1981) showed an example of the probabilistic approach which was applied to dynamic model prediction. It is also necessary that the primary factors affecting each parameter and their quantitative effects be clarified. With such quantitative information, the parameter values obtained under different conditions can be adjusted to other cases.

Despite the fact that the parameters have large uncertainties, general values have been used in some past evaluations. For example, the NRPB R-89 model (Simmonds et al., 1979) calculated the rate constant of root uptake from published T_f values which are applicable only in an equilibrium state. Some of the other transfer rates seem to be derived from the equilibrium-state parameters. We need more efforts to measure *in situ* transfer rates to establish a real dynamic model; the rates might be given as functions of environmental factors such as temperature, soil properties, and competing ion concentrations.

Some parameters used in previous models are insufficient in terms of their physical meanings, in other words, the mechanistic background. As a result, it has been difficult to predict quantitatively their variation due to environmental condition changes, which leads to large uncertainty in the results obtained. We should give physical descriptions to these parameters. A good example of such a description is seen in the calculation of downward infiltration rate (λ_i) (Hoffman and Baes, 1979) as follows:

$$\lambda_i = \frac{v_w}{d_s \cdot \left(1 + \frac{\rho_b}{\theta} \cdot K_d\right)} \qquad (20)$$

where v_w is the water percolation velocity in the soil [m day^{-1}]; d_s, the depth of the surface soil layer [m]; ρ_b, the bulk soil density [kg m^{-3}]; θ, the volumetric soil water content [L m^{-3}]; K_d, the soil-solution distribution coefficient [L kg^{-1}]. Almost the same descriptions for λ_i were presented by Muller and Prohl (1993) and Baes et al. (1984). Among the parameters in eq.20, the distribution

coefficient has a large uncertainty and is very difficult to determine *in situ* because, generally concentrations of radionuclides in actual soil solution are too low to be detected. Hence the K_ds have been obtained in laboratories using anthropogenic radiotracers. These experimental results, however, may be unsuitable for direct adaption to real fields where the environmental conditions are different.

The following sections will focus on how to reduce the uncertainty of two important, empirical parameters: soil-solution distribution coefficient and soil-to-plant transfer factor. Both are widely accepted parameters and a lot of values have already been reported (Ng and Hoffman, 1983; Keen and Frissel, 1983; Sheppard et al., 1984; Coughtrey et al., 1985; IUR, 1987; RWMC, 1988 and1990; Lembrechts and Koster, 1989). These values, however, show a large variation range and their variation characteristics are still unclear. Reliability in model predictions in a terrestrial ecosystem depends greatly on the accuracy of these parameters.

2.4 PARAMETER VALUES

The parameters involved in the environmental transfer models are considered to be specific to each case. We need to determine their proper values according to the data collected in real fields. Additionally, for increasing model reliability, accuracy and potential variability of each value adapted should be examined in a quantitative way using the accompanying environmental information. In many cases, however, the suitability of each parameter has not been sufficiently discussed. This section briefly outlines the features of two empirical parameters: K_d and T_f regarding their definitions, importance, and reference values.

2.4.1 Distribution coefficient (K_d)

A distribution coefficient, K_d, is one of the most important parameters in the environmental impact assessments for radioactive waste disposal (Inoue and Morisawa, 1976; Kato and Yabuta, 1986; Fukui, 1990). Migration of materials in soil is affected greatly by the interaction between soil solid and soil solution. The K_d can be an index which describes sorption, one of the most important reactions.

The K_d is defined as a concentration ratio between soil solid phase and liquid phase as follows:

$$K_d = \frac{C_l}{C_s} \qquad (21)$$

where C_l is the radionuclide concentration in soil solution [Bq g^{-1}] and C_s, that in soil solid phase [Bq g^{-1}]. The K_d, which has been empirically obtained in many cases, corresponds to reversible sorption, which normally means ion exchange. Adsorption of radionuclide onto soil solid surface by cation exchange is expressed as

$$\text{Soil} - B^+ + A^+ \rightarrow \text{Soil} - A^+ + B^+$$

where A^+ is the cationic radionuclide added into soil solution and B^+, the exchangeable cation previously adsorbed onto the soil surface. Electric capacity of soil surface for adsorbing cation is termed cation exchange capacity (CEC) [mmol(+) $100g^{-1}$-soil] which is given as the product of the charge density of soil surface and the specific surface area (Bolt and Bruggenwert, 1978). When the pH is within 5 to 7, the CEC is attributed mainly to the content of clay minerals having large surface charge density. The relationship between the K_d of ion *a* (K_{d_a}) and that of ion *b* (K_{d_b}) is expressed as the following equation called Kerr's equilibrium model (Bolt and Bruggenwert, 1978):

$$K_{d\_a} = K_{a/b} \cdot K_{d\_b} \qquad (22)$$

where $K_{a/b}$ [-] is referred to as a selectivity coefficient. The $K_{a/b}$ is considered to be affected by the ion composition in soil solution. The above equation suggests that we can estimate radionuclide K_ds from the concentration of major elements.

Being very convenient for use, the K_d has been widely applied to environmental impact assessments (NCRP, 1984). Already many K_ds have been reported (Sheppard et al., 1984; Coughtrey et al., 1985; RWMC, 1990). The value ranges of reference K_ds are summarized in **Table 1** for selected nuclides (Mn, Co, Zn, Sr, I, Cs, Ra, U, Pu). As shown in the table, the K_ds have large variations up to 5 orders. Though some statistical information on the variability of the K_ds has been reported for selected radionuclides (Baes et al., 1984; Sheppard et al., 1984), estimating their appropriate variation range in an actual field is still a difficult task.

Most K_ds have been obtained in laboratory experiments since radionuclides and

Table 1. Variation range of reference K_ds for selected nuclides.

nuclide	Foreign countries[a]			Japan[b]		
	Sand	Silt	Clay	Sand	Loam	Clay
Mn	–	–	–	4.0×10^{0}–1.2×10^{4}	–	7.0×10^{1}–1.0×10^{4}
Co	–	–	–	3.0×10^{0}–4.2×10^{3}	1.0×10^{1}–3.3×10^{3}	2.0×10^{1}–1.2×10^{4}
Zn	1.0×10^{-1}–2.1×10^{3}(5)	3.6×10^{0}–1.0×10^{2}(2)	–	2.0×10^{1}–8.0×10^{3}	–	2.0×10^{2}–4.0×10^{3}
Sr	2.0×10^{0}–1.1×10^{2}(26)	8.0×10^{0}–3.0×10^{2}(20)	8.0×10^{0}–1.2×10^{3}(6)	5.0×10^{-1}–6.8×10^{2}	5.0×10^{-1}–2.7×10^{3}	1.0×10^{1}–9.0×10^{3}
I	2.0×10^{-1}–1.2×10^{0}(7)	1.8×10^{-1}–1.5×10^{0}(11)	1.0×10^{0}–1.8×10^{0}(4)	4.0×10^{1}	–	–
Cs	1.0×10^{1}–1.0×10^{4}(24)	6.5×10^{2}–3.0×10^{4}(20)	6.5×10^{1}–3.2×10^{4}(5)	3.0×10^{0}–8.0×10^{3}	1.0×10^{0}–2.5×10^{3}	5.0×10^{1}–7.0×10^{4}
Ra	1.1×10^{2}–3.8×10^{4}(3)	2.0×10^{4}–9.5×10^{5}(4)	7.0×10^{2}–5.6×10^{4}(8)	–	–	–
U	1.3×10^{-1}–1.6×10^{1}(2)	–	2.0×10^{2}–7.9×10^{5}(3)	4.6×10^{1}	–	–
Pu	3.3×10^{1}–6.9×10^{3}(19)	2.3×10^{2}–9.0×10^{4}(8)	3.2×10^{2}–1.9×10^{5}(13)	9.7×10^{2}–1.9×10^{3}	2.4×10^{3}–4.6×10^{3}	6.0×10^{2}–7.0×10^{2}

[a] From Sheppard et al.(1984); the number of samples is indicated in parentheses.

[b] From RWMC (1990).

other toxic substances are limited for use as a tracer in field experiments. The methods for measurement of K_d can be divided into two types: batch and column techniques. Typically in the batch technique, soil and solution including the pollutant are kept in contact for a sufficient period to reach equilibrium (several days), while being shaken, before measuring the concentration in the supernatant for the calculation of K_d. In the column technique, the K_d is calculated by comparing the breakthrough curve with that of the water (e.g. Inoue and Kaufman, 1963). The batch technique has been widely used since it is simpler and generally faster than the column technique. However, the column technique is considered to give more realistic results because the sollutes in the soil solution move depending on the matric potential gradient. The K_d obtained by the batch technique tends to be larger than that by using the column (Inoue and Morisawa, 1976; Relyea et al., 1980). In both methods, however, it should be noted that the K_d value obtained depends greatly on the experimental conditions (Rancon, 1986; Fukui, 1990; RWMC, 1990; Yasuda et al., 1992; 1993) such as competing ion concentration, soil properties, temperature, oxidation-reduction potential, and amount of organic materials. Additionally, the measurement techniques such as solution volume, contacting time, and initial concentration also affect the K_ds (IAEA, 1985).

Because the effects of the factors listed above are complicated and interrelated, it has still been difficult to evaluate the suitability of the K_d adapted. The standard method to determine *in situ* K_d has been one of the important subjects for reliable model predictions. In particular, little information on K_ds is available for agricultural soils (RWMC, 1990). We need further effort to decrease the uncertainty of the K_ds by collecting accompanying information.

2.4.2 Soil-to-plant transfer factor (T_f)

In the field of radioecology, the amount of radionuclides transferred from soil to plant is calculated with a transfer factor (T_f). Because of its convenience for use, the T_f has been widely applied to environmental impact assessments in the evaluation of internal exposure through soil-crop pathway (ICRP, 1978; Morisawa et al., 1985). The T_f is defined as a concentration ratio between crop edible parts and soil as follows:

$$T_f = \frac{C_{crop}}{C_{soil}} \qquad (23)$$

where C_{crop} is the radionuclide concentration in crop edible parts [Bq g^{-1}]; and C_{soil}, that in soil [Bq g^{-1}]. Many T_fs have been reported on a dry or wet crop weight basis (Menzel, 1965; USNRC, 1977; IAEA, 1982; Ng et al., 1982; IUR, 1987). Large variations ran up to 6 orders have been found for those T_fs (Ng and Hoffman, 1983; Keen and Frissel, 1983; NCRP, 1984). In **Table 2**, the value ranges of some reference T_fs obtained in foreign countries (Ng et al., 1982; NCRP, 1984) and those in Japan (RWMC, 1988) are summarized.

The T_f vary greatly depending on changes of the growth conditions such as the crop species (Keen and Frissel, 1983; Ng and Hoffman, 1983; Uchida et al., 1987; RIVM, 1988), soil properties (Ng et al., 1979c; Ng and Hoffman, 1983; Yasuda et al., 1992), competing ion concentration (Kuhn et al., 1984; Schuller et al., 1988; Shaw and Bell, 1991; Yasuda and Inoue, 1992; Shaw, 1993), and chemical form of the element (Muramatsu et al., 1983; Whitehead, 1973). Additionally, the difference of measurement techniques -- pot or lysimeter experiment or field survey -- is considered to cause the variation of T_f (Keen and Frissel, 1983). For reliable model predictions, the variational characteristics of T_fs related to the growth conditions should be clarified.

Table 2. Variation range of reference T_fs on the wet weight basis for selected nuclides.

nuclide	Foreign country[a]	Japan[b]
Mn	6.9×10^{-3}-3.4×10^{0}	1.0×10^{-3}-2.0×10^{0}
Co	2.2×10^{-3}-3.0×10^{-1}	2.0×10^{-4}-3.0×10^{-1}
Zn	7.2×10^{-3}-1.6×10^{0}	3.0×10^{-2}-2.0×10^{0}
Sr	1.6×10^{-3}-1.7×10^{0}	4.0×10^{-3}-1.0×10^{0}
I	2.0×10^{-4}-1.2×10^{-1}	1.0×10^{-4}-8.0×10^{-3}
Cs	1.5×10^{-5}-2.9×10^{-1}	3.0×10^{-4}-8.0×10^{-1}
Ra	1.1×10^{-4}-2.0×10^{-1}	2.0×10^{-4}-2.0×10^{0}
U	1.7×10^{-4}-9.0×10^{-3}	4.0×10^{-5}-1.0×10^{-2}
Pu	1.2×10^{-7}-2.0×10^{-2}	9.0×10^{-6}-2.0×10^{-1}

[a] From Ng et al. (1982) and NCRP (1984).
[b] From RWMC (1988).

Table 3. Default T_fs for selected nuclides.

nuclide	USNRC (1977)	IAEA (1982)	
	crops[a]	crops[a]	forage plants[b]
Mn	2.9×10^{-2}	5×10^{-1}	3×10^{-1}
Co	9.4×10^{-3}	3×10^{-2}	4×10^{-1}
Zn	4.0×10^{-1}	4×10^{-1}	5×10^{-1}
Sr	1.7×10^{-2}	3×10^{-1}	2×10^{0}
I	2.0×10^{-2}	2×10^{-2}	9×10^{-1}
Cs	1.0×10^{-2}	3×10^{-2}	1×10^{-1}
Ra	–	4×10^{-2}	2×10^{-1}
U	–	2×10^{-3}	1×10^{-2}
Pu	–	5×10^{-4}	1×10^{-3}

[a] On a wet weight basis [Bq g^{-1} wet vegetation per Bq g^{-1} dry soil].
[b] On a dry weight basis [Bq g^{-1} dry vegetation per Bq g^{-1} dry soil].

Some statistical information on T_f variation has been supplied (Ng et al, 1979c; Ng et al., 1982; Keen and Frissel, 1983; Ng and Hoffman, 1983; Yasuda and Uchida, 1993d). In two of these reports, it was pointed out that classification of soil type seems to be effective for decreasing the uncertainty of T_f (Ng and Hoffman, 1983; Yasuda and Uchida, 1993c). The statistical approach, however, requires many T_fs collected in field surveys. In general cases, T_f data are insufficient for statistical analyses by which the suitability and the potential variability of the T_f can be evaluated. Though some default T_fs have been stated for major nuclides (USNRC, 1977; IAEA, 1982) as shown in **Table 3**, it should be noted that these default values may cause potential errors. If the quantitative relationship between T_f and each factor is known, the uncertainty of T_f can be reduced by utilizing information on the growth conditions. Peterson (1983) presented a representative T_f for each crop species (**Table 4**). Such efforts are critical for reliability improvement in internal dose evaluation.

2.4.3 Other parameters

Some values of selected model parameters used in the previous transfer models (USNRC, 1977; Hoffman and Baes, 1979; NCRP, 1984) are shown in **Table 5**.

Table 4. Representative T_fs obtained on a dry weight basis (Peterson, 1983); these are the 84th percentile bound on the mean and the number of samples is indicated in parentheses.

nuclide	Forage		Grain		Leaf	Root		Legumes	Fruits
	Legume	Grasses	Corn Rice	Wheat etc.	Cabbage Spinach, etc.	Potato	Radish Carrot, etc.	Bean, Pea, etc.	Tomato, Apple, etc.
^{54}Mn	–	5.3×10^{-1} (2)	–	3.5×10^{-1} (2)	7.2×10^{-1} (1)	1.4×10^{-1} (1)	8.0×10^{-2} (4)	2.4×10^{-1} (1)	4.6×10^{-2} (4)
^{60}Co	–	4.0×10^{-2} (8)	1.0×10^{-2} (1)	2.9×10^{-2} (3)	8.0×10^{-2} (6)	4.6×10^{-2} (3)	4.5×10^{-2} (3)	–	–
^{65}Zn	–	–	–	–	6.8×10^{-1} (6)	–	–	–	–
^{90}Sr	3.1×10^{-0} (15)	1.3×10^{-1} (4)	2.7×10^{-2} (7)	2.2×10^{-1} (25)	2.2×10^{-0} (12)	4.7×10^{-1} (5)	1.8×10^{-0}(17)	1.8×10^{-0} (6)	2.4×10^{-1} (7)
^{129}I	1.8×10^{-3} (1)	2.5×10^{-1} (1)	–	–	–	3.0×10^{-2} (1)	3.0×10^{-2} (4)	–	–
^{137}Cs	9.3×10^{-2} (9)	4.8×10^{-2} (24)	7.8×10^{-3}(3)	1.9×10^{-2} (11)	2.2×10^{-2} (14)	3.2×10^{-2} (3)	3.7×10^{-2} (11)	6.1×10^{-2} (7)	2.6×10^{-2} (4)
^{226}Ra	1.0×10^{-1} (24)	4.0×10^{-2} (9)	–	5.8×10^{-2} (4)	4.4×10^{-1} (2)	–	1.7×10^{-2} (2)	3.5×10^{-3} (1)	–
^{238}U	3.9×10^{-4} (1)	5.0×10^{-3}(11)	–	1.6×10^{-4} (2)	–	9.0×10^{-4} (1)	3.0×10^{-4} (1)	1.0×10^{-3} (2)	1.7×10^{-3} (3)
^{239}Pu	2.3×10^{-4} (24)	9.2×10^{-5} (35)	–	1.5×10^{-6} (20)	1.75×10^{-4}(3)	1.4×10^{-3}(1)	3.7×10^{-4} (5)	8.1×10^{-6} (19)	–

Table 5. Values of other selected parameters used for model prediction in a terrestrial ecosystem.

parameter	value		unit	reference
Half-life of radioactive decay (T_H)	^{60}Co :	5.27	y	IAEA, 1986a
	^{90}Sr :	28.8	y	
	^{131}I :	8.3×10^{-3}	y	
	^{137}Cs :	30.0	y	
	^{239}Pu :	2.41×10^{4}	y	
Water infiltration rate (v_w)	36.5-376		cm/y	Hoffman and Baes, 1979
Soil water content by weight (θ)	0.03-0.40		mL/cm^3	
Annual crop yield per unit area (Y_c)	Pasture	0.7	kg-wet $/m^2/y$	USNRC, 1977
	Leafy vegetables	2.0	kg-wet $/m^2/y$	
Areal surface soil density (W_{soil})	224		kg/m^2	Baker et al., 1976
	240		kg/m^2	USNRC, 1977

These are half-life of radioactive decay (T_H), water infiltration velocity (V_h), volumetric soil water content (θ), annual crop yield per unit area (Y_c), and areal surface soil density (W_{soil}). IAEA (1994) has published a report which summarizes the parameters such as deposition-retention (interception) coefficients and translocation coefficients in inner parts of crops in temperate regions. As is the same with the K_d and T_f, these parameters (except for T_H) also have some variabilities due to changes of geological and meteorological conditions. We need to collect more site-specific data and to clarify their potential variabilities based on the frequency of those observations.

CHAPTER 3. ACCURACY IMPROVEMENT OF DISTRIBUTION COEFFICIENT

3.1 INTRODUCTION

Many K_ds have been already obtained as shown in Table 1. However, as mentioned in section 2.4.1, applying these reference K_ds to a real assessment would cause a large uncertainty of the results obtained. For improving the reliability of the K_d, we need to collect pertinent information on its variational characteristics due to environmental condition changes.

In this chapter, the K_ds of five cationic radionuclides were measured by the batch technique for agricultural soils collected in Japan. The radionuclides were: ^{54}Mn (half life: 312 d), ^{60}Co (5.27 y), ^{65}Zn (244 d), ^{85}Sr (28.8 y), and ^{137}Cs (30.0 y). All of them were in a carrier-free state. The ^{90}Sr and ^{137}Cs are of primary importance in evaluating human exposure when a nuclear accident occurs; the other three radionuclides (^{54}Mn, ^{60}Co, ^{65}Zn) are important as activated products (UNSCEAR, 1982). Stable isotopes of Mn, Co, and Zn are considered to have a relatively high potential to contaminate agricultural soils because they are contained in large amounts in industrial wastes (Martin and Coughtrey, 1982).

3.2 EXPERIMENTAL STUDIES

3.2.1 Effect of temparature

Materials and methods. Chronological changes of the radionuclide K_ds under different temperatures were examined for an agricultural soil taken from Tokai, Ibaraki, Japan. The temperatures were set to three levels: 3°C, 13°C, and 23°C. The soil was dried at room temperature for about one month and sieved to 2 mm. The physico-chemical properties of the soil are shown in **Table 6**. The water content was measured by drying the soil at 110 °C for about 5 hours. The cation exchange capacity (CEC), exchangeable potassium (ex.K), and exchangeable calcium (ex.Ca) were measured by the established method (Schollenberger and Simon, 1945). Soil was digested in a strong acid (60% HNO_3 15mL + 48% H_2SO_4

Table 6. Physico-chemical properties of the sample soil on a dry weight basis.

water content (air-dried) [mL/g]	pH (H_2O)	CEC [mmol(+)/100g-soil]	particle			elementary composition			
			density [g/cm^3]	effective diameter[a] [mm]	uniformity coefficient[b]	exchangeable K [mmol/g]	exchangeable Ca [mmol/g]	total Al [mmol/g]	total C [mmol/g]
0.082	5.3	16.6	2.06	0.014	18	3.0×10^{-3}	1.2×10^{-2}	4.3	3.6

[a] The 10 percentile diameter.
[b] The ratio of 60 percentile diameter to the effective diameter.

Table 7. Chemical properties of the rainwater (contact solution).

pH	EC [mS/cm]	Na^+ [mmol/L]	NH_4^+ [mmol/L]	K^+ [mmol/L]	Mg^{2+} [mmol/L]	Ca^{2+} [mmol/L]
4.5	0.065	0.327	0.096	0.013	0.041	0.044

4mL + 46% HF 5mL) before measurement of total aluminum content (Al) (Geological Survey of Japan, 1980). Potassium concentration was determined with an atomic absorption spectrometer (Shimadzu, AA646); calcium and aluminum, with ICP (Seiko, SPS1100H); total carbon (C), with a CN corder (Yanagimoto, MT-3) (Dojoyoubun-bunsekihou iinkai, 1991).

The contact solution was rainwater collected with a stainless steel bottle placed outdoors in Tokai. The solution was filtered through a 0.45 μm membrane filter and kept in a refrigerator until the beginning of the experiments. The chemical properties of the contact solution are shown in **Table 7**. In measurement of K_d, to 50 mL polyethylene bottles were added 3 g of soil, and 30 mL of contact solution which contained 1-10 kBq each of the five radionuclides. Bottles were shaken at 150 reciprocations per minutes for 216 hours while keeping the temperature constant. Supernatant solution (5 mL) of each sample was collected at 24, 96, and 216 hours. The supernatant was centrifuged at 4,000 rpm and then filtered through a 0.45 μm membrane filter before for γ-ray intensity measurement with a Ge-detector (Ortec GEM-30185) coupled to a multichannel analyzer (Seiko, EG&G 7800). The pH and the electrical conductivity (EC) [mS/cm] in the supernatant were also measured with a pH meter (Horiba, F-13) and an EC meter (Yokogawa, SC82), respectively.

The K_d [L kg^{-1}] of each radionuclide was calculated by

$$K_d = \frac{(C_i - C_l)\cdot W_l}{C_l \cdot W_g} \qquad (24)$$

where C_i is the initial concentration of a radionuclide in the solution [Bq L^{-1}]; C_l, the concentration of a radionuclide in the supernatant solution after shaking with soil [Bq L^{-1}]; W_l, the solution volume [L]; and W_g, the soil weight [kg]. The way of calculating K_d in other sections is the same with eq.24.

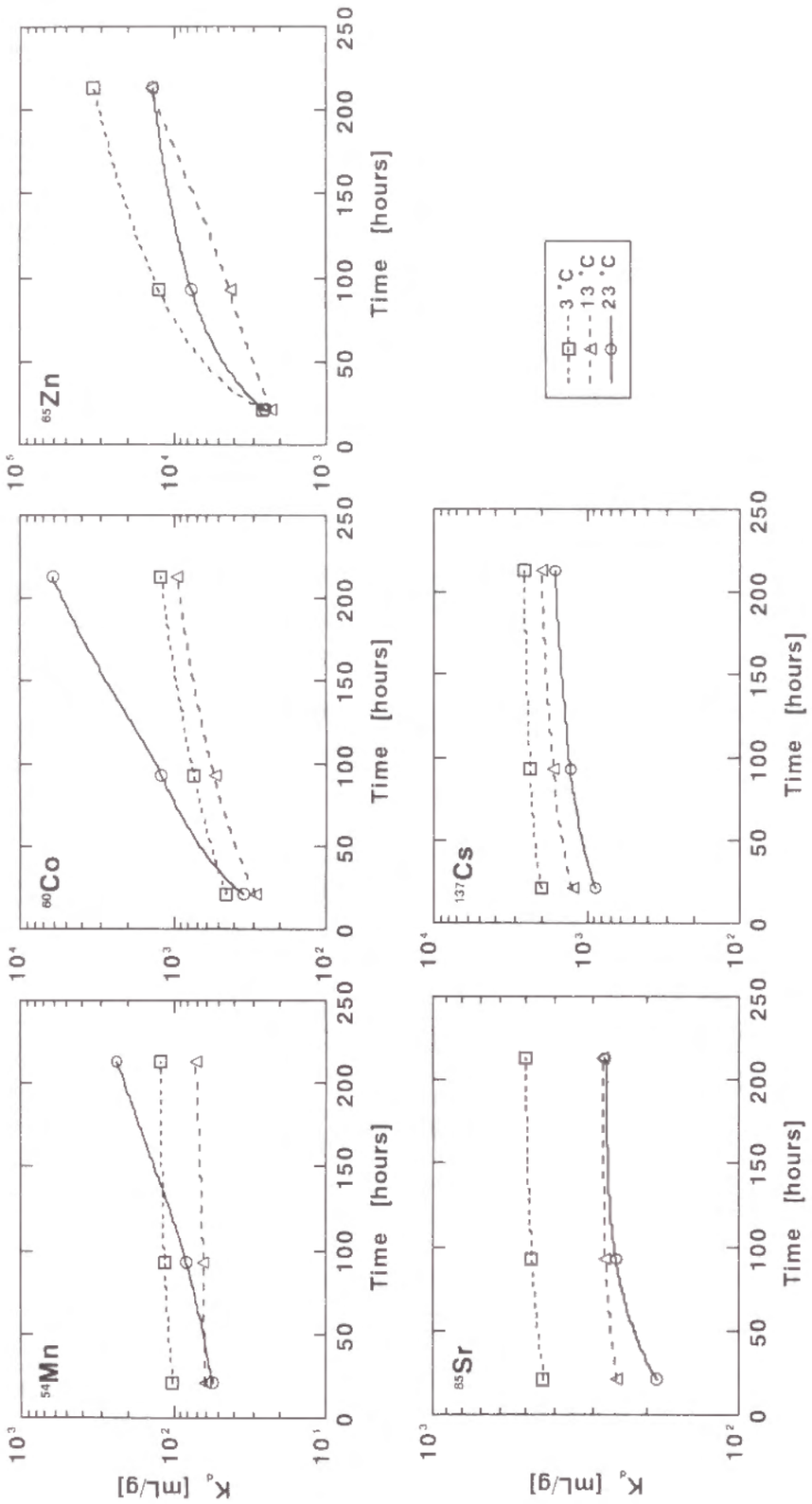

Figure 12. Variation of the radionuclide K_ds with time under different temperatures.

Results and discussion. The K_d variations of the five radionuclides with time are shown in **Figure 12**. For all radionuclides, thermal effects are found. For ^{60}Co and ^{54}Mn, the K_ds are still increasing at 216 hours at 23 °C while they reach equilibrium smoothly at 3°C and 13°C. Because the pH and the EC in the supernatant do not change much (not shown here), such an increase of apparent sorption of these radionuclides is probably due to microbial effects. For the other three radionuclides (^{85}Sr, ^{137}Cs, and ^{65}Zn), their K_ds at 3°C are larger than those at 13°C and 23°C; their K_d values become nearly stable after 96 hours.

3.2.2 Effect of competing ions

Materials and methods. To investigate the effects of competing ion concentrations, four solutions having different ion concentrations were left in contact with the soil (the same as in section 3.2.1). Nutrient solution was used because the elemental composition of the solution in agricultural soil where crops are grown is assumed to be similar to it. The four solutions were the standard nutrient solution and solutions diluted to 1/2, 1/5, and 1/10 of the standard solution. The chemical properties of the standard nutrient solution are shown in **Table 8**. The Ca and K were given as nitrate ($Ca(NO_3)_2 \cdot 4H_2O$ and KNO_3); Mg, as sulfate ($MgSO_4 \cdot 7H_2O$); and N, as phosphate ($NH_4H_2PO_4$). Each sample, 10 g soil and 25 mL nutrient contact solution in a 50 mL polyethylene bottle, was shaken at 100 rpm with a shaker which being kept at 23 °C. After shaking for 1, 24, 72, and 168 hours, an aliquot of the supernatant solution (5 mL) was centrifuged at 4,000 rpm, filtered through a 0.45μm membrane filter, and analyzed for γ-ray intensity; the detailed procedures are the same as those stated in section 3.2.1. Each treatment was repeated three times.

Results and discussion. The changes of radionuclide K_ds are shown in **Figure 13**. Each K_d have become 10 to 20 fold smaller with increased co-existing ion

Table 8. Chemical properties of the standard nutrient solution; solutions having 1/2, 1/5, and 1/10 of these values were also used in the experiment.

pH	EC [mS/cm]	Na^+ [mmol/L]	NH_4^+ [mmol/L]	K^+ [mmol/L]	Mg^{2+} [mmol/L]	Ca^{2+} [mmol/L]
4.7	2.4	0.07	1.7	7.8	1.9	3.6

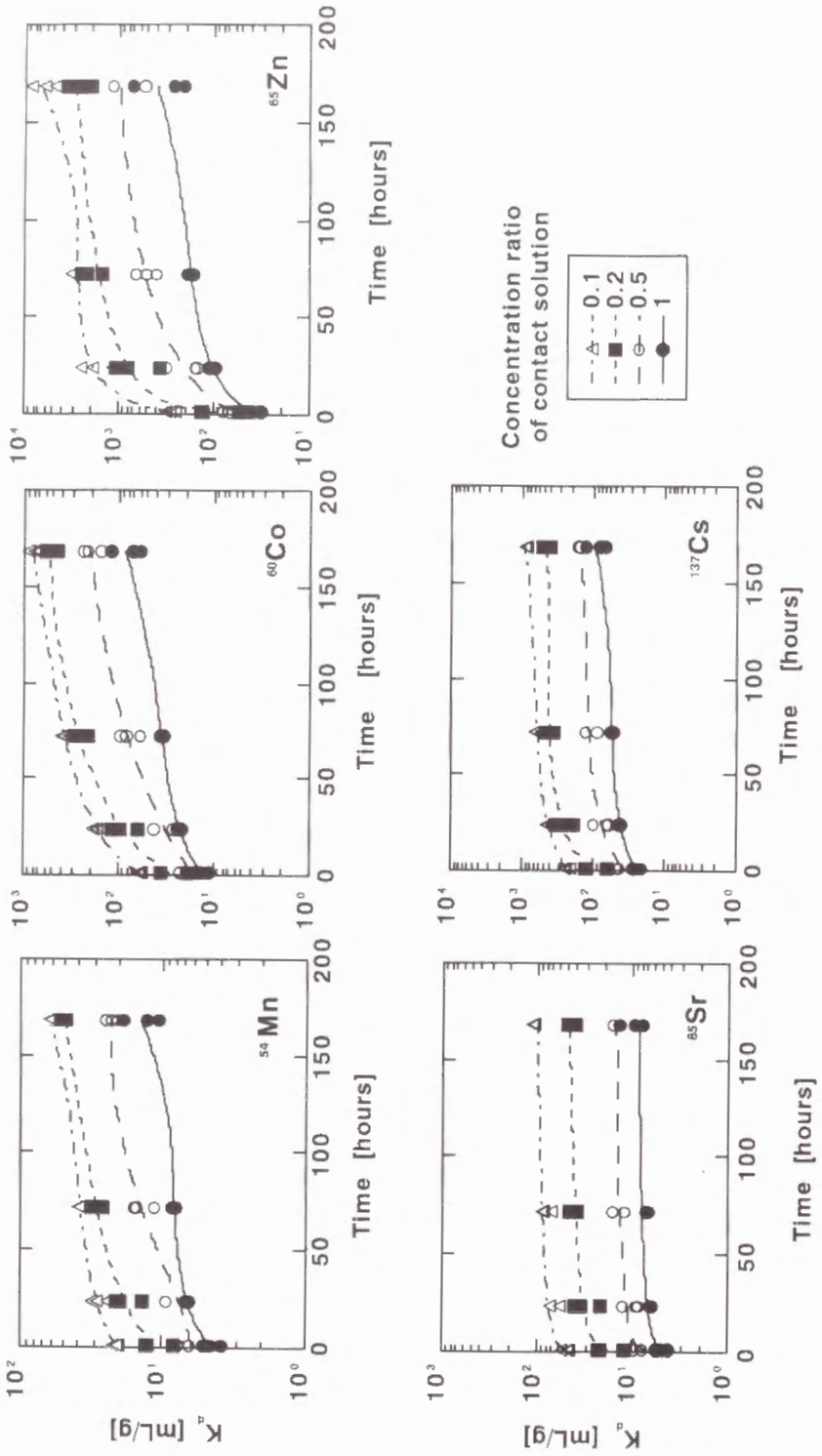

Figure 13. Variation of the radionuclide K_ds with time for different nutrient solutions; the value in the legend is the concentration ratio of the contact solution to the original solution shown in Table 8.

concentration. This indicates that the K_ds of all radionuclides are affected by the competing ion concentration, that is, these radionuclides compete with other ions in adsorption to the soil. For ^{60}Co, ^{54}Mn and ^{65}Zn, the K_ds are still increasing at 168 hours; possibly microbial effects were operating in their sorption processes. For the other two radionuclides (^{85}Sr and ^{137}Cs), their K_d values become nearly stable after 72 hours.

3.2.3 Effect of soil properties

Materials and methods. To discuss effects of soil properties, the K_ds were measured for thirty-six agricultural soils (18 upland soils and 18 paddy soils) collected from the surface (0-20cm depth) throughout Japan. The soils were dried at room temperature for about one month and sieved to 2 mm. The contact solution used was the rainwater shown in Table 7. All soils were analyzed for 12 soil properties: water content [L/kg], pH(H_2O), cation exchange capacity (CEC) [mmol(+) kg^{-1}], exchangeable potassium content (ex.K) [mmol kg^{-1}], exchangeable calcium content (ex.Ca) [mmol kg^{-1}], total calcium content (total Ca) [mmol kg^{-1}], total potassium content (total K) [mmol kg^{-1}], total aluminum content (total Al) [mmol kg^{-1}], total iron content (total Fe) [mmol kg^{-1}], ammonium productivity (NH_4-pr.) [mmol kg^{-1}], total carbon content (total C) [g kg^{-1}], and total nitrogen content (total N) [g kg^{-1}]. For pH (H_2O), the soil was shaken with a soil/water ratio of 1:2.5 (Jpn. Soc. Soil Sci. & Plant Nutr., 1988). Water content, CEC, ex.K, ex.Ca, total Al, and total C were measured by the methods mentioned in section 3.2.1. Before measurement of total ions (K, Ca, Fe, and Al), the sample soil was decomposed in a strong acid (HNO_3 + H_2SO_4 + HF) (Geological Survey of Japan, 1980). The Fe was measured with an inductively coupled plasma spectrometer (Seiko, SPS1100H). NH_4-pr. was defined as the ammonium amount germinated from the soil maintained at 30°C for a period of 28 days. The C and N were analyzed with a reference type thermal conductivity detector (Yanagimoto, CHN corder MT-3) (Dojoyoubun-bunsekihou-iinkai, 1991).

Bottles containing 3 g soil and 30 mL contact solution were shaken at 150 rpm for 7 days while keeping the temperature at 23°C. After 168 hours, a supernatant aliquot (5 mL) of each sample was centrifuged, filtered through a 0.45μm membrane filter, and analyzed for γ-ray intensity. The pH and the EC in the supernatant were also measured.

Distribution of soil properties. Major statistical values (minimum, median, maximum, mean, and standard deviation) as to the soil properties are shown in **Table 9**. To discuss their variational characteristics, the probability distribution type of each soil property was examined by the Shapiro-Wilk test (Shapiro and Wilk, 1965); normal distribution and log-normal distribution were tested. A normal type distribution is found for three properties: pH(H_2O), total Ca, and total K. The other 9 properties show log-normal type distributions. For CEC, ex.Ca, total Ca, C, and N, 90% (5-95 percentile) samples were used in this test because they have 1 or 2 outlier values. In comparison of each soil property between upland and paddy soils by the t-test, no significant difference is found ($p<0.01$) except for NH_4-pr. (**Table 9**).

Distribution of radionuclide K_d. The probability distributions of the K_ds are shown in **Figure 14**. From the Shapiro-Wilk test (Shapiro and Wilk, 1965) using 90% (5-95 percentile) samples, the K_d distributions for ^{60}Co, ^{65}Zn, ^{85}Sr, and ^{137}Cs are judged as not normal-types ($p<0.02$), but log-normal types ($p>0.18$). For ^{54}Mn, its K_d distribution is neither a normal nor a log-normal type ($p<0.01$); the slope of its distribution curve is bent near the 50 percentile value.

The statistical values of K_ds are shown in **Table 10**. The variation of K_d is up to two or three orders; the ratio of max to min is about 200 for ^{137}Cs, but 2000 for ^{54}Mn. Their geometric means are: 1.8×10^3 [L kg^{-1}] for ^{60}Co, 2.0×10^3 [L kg^{-1}] for ^{65}Zn, 1.7×10^2 [L kg^{-1}] for ^{85}Sr, and 1.8×10^3 [L kg^{-1}] for ^{137}Cs. In comparison of each geometric mean between upland and paddy soils by the t-test, no significant difference is found ($p>0.05$). Actually, however, *in situ* K_d would possibly show some difference between upland and paddy soils because the conditions such as oxidation-reduction potential might not be the same. The t-test was not adopted for ^{54}Mn because the type of its K_d distribution was unclear .

Correlations with soil properties. Spearman's rank correlation coefficients between the K_d and the soil properties are summarized in **Table 11**. Adequate correlations (Spearman correl. coeff.>0.6) are found in the following combinations:

^{54}Mn - ex.Ca

^{65}Zn - water cont.

Table 9. Statistical characteristics of soil properties.

soil property	distribution type [a]	min	median	max	(geometric) mean [b] all (n=36)	(geometric) mean [b] upland (n=18)	(geometric) mean [b] paddy (n=18)	std. dev. (com. log.) [b] all (n=36)	std. dev. (com. log.) [b] upland (n=18)	std. dev. (com. log.) [b] paddy (n=18)	p value in t-test [c]
water cont. [L/kg]	log-normal	0.003	0.025	0.102	0.025	0.024	0.025	0.36	0.44	0.27	0.85
pH (H_2O)	normal	2.98	5.81	7.43	5.85	5.85	5.85	0.82	0.99	0.63	0.99
CEC [mmol(+)/kg]	log-normal	14	131	390	121	107	137	0.30	0.36	0.21	0.28
ex.Ca [mmol/kg]	log-normal	3.3	34.3	194.6	34.5	29.0	41.0	0.34	0.42	0.24	0.19
ex.K [mmol/kg]	log-normal	0.3	2.2	16.4	2.5	2.5	2.4	0.35	0.41	0.29	0.85
total Ca [mmol/kg]	normal	14	265	714	293	322	265	202	224	179	0.40
total K [mmol/kg]	normal	19	230	545	240	205	275	149	155	138	0.16
total Fe [mmol/kg]	log-normal	281	693	2006	667	679	653	0.20	0.23	0.19	0.80
total Al [mmol/kg]	log-normal	2405	3180	4418	3192	3214	3170	0.06	0.08	0.04	0.77
NH_4-pr. [mmol/kg]	log-normal	0.39	2.44	9.44	1.94	1.23	3.05	0.36	0.35	0.24	0.00
total C [g/kg]	log-normal	0.7	17.0	102.1	15.6	13.1	18.6	0.45	0.56	0.29	0.32
total N [g/kg]	log-normal	0.1	1.6	7.2	1.5	1.3	1.8	0.38	0.48	0.24	0.24

[a] Each distribution type was determined by the Shapiro-Wilk test with a significance level of $p>0.05$; for CEC, ex.Ca, total Ca, C, and N, 90% (5-95 percentile) samples were used in this test.

[b] The arithmetic means and the standard deviations are shown for normal-type distributions and the geometric means and the standard deviations of the common logarithms for log-normal type distributions.

[c] This is the probability when the (geometric) means were assumed to be the same between the upland and paddy soils.

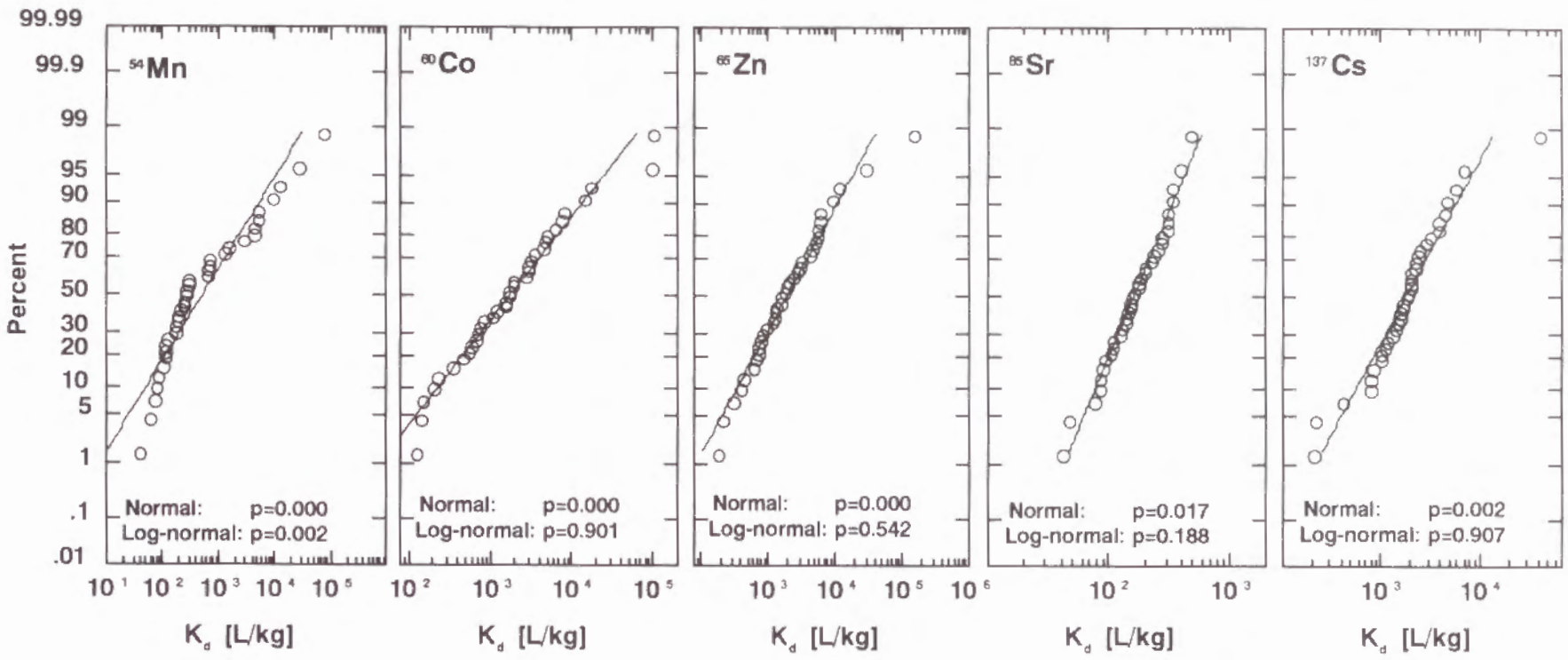

Figure 14. Probability distributions of the radionuclide K_ds; p value in the figure is a probability obtained in the Shapiro-Wilk test using 90 % (5-95 percentile) data.

Table 10. Statistical characteristics of the obtained K_ds.

Nuclide	distribution type [a]	min	median	max	geometric mean			std. dev. com. log.			p value in t-test [b]
					all (n=36)	upland (n=18)	paddy (n=18)	all (n=36)	upland (n=18)	paddy (n=18)	
^{54}Mn	-	39.7	271	79040	-	-	-	-	-	-	-
^{60}Co	log-normal	130	1735	103600	1837	1914	1762	0.70	0.64	0.78	0.88
^{65}Zn	log-normal	185	1756	153100	2027	2317	1770	0.60	0.59	0.61	0.56
^{85}Sr	log-normal	42.0	163	498	166	156	177	0.25	0.29	0.21	0.51
^{137}Cs	log-normal	215	1896	42610	1849	2109	1626	0.41	0.50	0.31	0.42

[a] Each distribution type was determined by the Shapiro-Wilk test with a significance level of $p>0.05$.

[b] This is the probability when the geometric means were assumed to be the same between upland and paddy soils.

Table 11. Spearman's rank correlation coefficients between the soil properties and the radionuclide K_ds.

nuclide	water cont.	pH	CEC	ex. Ca	ex. K	total Ca	total K	total Fe	total Al	NH_4-pr.	Total C	Total N
^{54}Mn	0.55	0.57	0.58	0.75	0.48	0.23	-0.13	0.22	0.31	-0.18	0.40	0.20
^{60}Co	0.53	0.53	0.52	0.59	0.31	0.21	-0.15	0.27	0.47	-0.27	0.37	0.11
^{65}Zn	0.67	0.28	0.43	0.27	-0.05	0.52	-0.46	0.59	0.47	-0.41	0.26	0.15
^{85}Sr	0.57	-0.02	0.64	0.35	0.09	0.37	-0.34	0.54	0.51	0.10	0.54	0.43
^{137}Cs	0.12	0.18	0.12	0.19	0.44	0.22	0.04	0.20	0.24	-0.21	-0.20	-0.25

^{85}Sr - CEC.

Good correlations (Spearman correl. coeff.>0.5) are found in:

^{54}Mn - water cont., pH(H_2O), CEC

^{60}Co - water cont., pH(H_2O), CEC, ex.Ca

^{65}Zn - total Ca, Fe

^{85}Sr - water cont., total Al, Fe, C.

Among these properties, ex.Ca and CEC are considered to represent the ability of cation exchange (Wahlberg, et al., 1965). Contents of C is showing an amount of organic materials which have the groups absorbing ions (Kabata-Pendias and Pendias, 1984). Water cont. and Al possibly relate to the clay content, i.e. the sorption area (Ahlrichs, 1972). The sorption features of ^{65}Zn , of which K_d correlates to total Ca, seem to be different from those of the other four radionuclides. Though all soil properties have little correlation with the K_d of ^{137}Cs, a relatively high correlation (Spearman correl. coeff. =0.44) is found with ex.K.

The relationships between the K_d and the soil property showing the highest correlation for each radionuclide are plotted in **Figure 15**. The R is Pearson's correlation coefficient between their logarithms; R_{all} is that calculated for all data and $R_{5\text{-}95\%}$, for 90% (5-95 percentile) data. Good correlations (R=0.5-0.7) are seen for all combinations. A particularly adequate correlation is seen between the CEC and the K_d of ^{85}Sr; this can be explained theoretically by an ion exchange mechanism (Wahlberg, et al., 1965). Though the relationships obtained here possibly change due to some outlier values, these results suggest that the K_ds of selected radionuclides can be determined more precisely by being classified according to the soil properties.

Spearman's rank correlation coefficients between the K_d and the solution properties are summarized in **Table 12**. The pH(H_2O) and supernatant pH also correlate with the K_ds of ^{54}Mn and ^{60}Co. Their apparent sorption might be attributed to changes of their chemical forms according to pH (Lindsay, 1972; Lindsay, 1979). The K_ds of both radionuclides also have a good correlation with ex.Ca and relatively high correlation with supernatant EC; these findings are probably due to the results of ion exchange, that is, an increase of Ca^{2+} dissolved by exchange with H^+. More Ca^{2+} is considered to be exchanged for the soil having higher contents of ex.Ca.

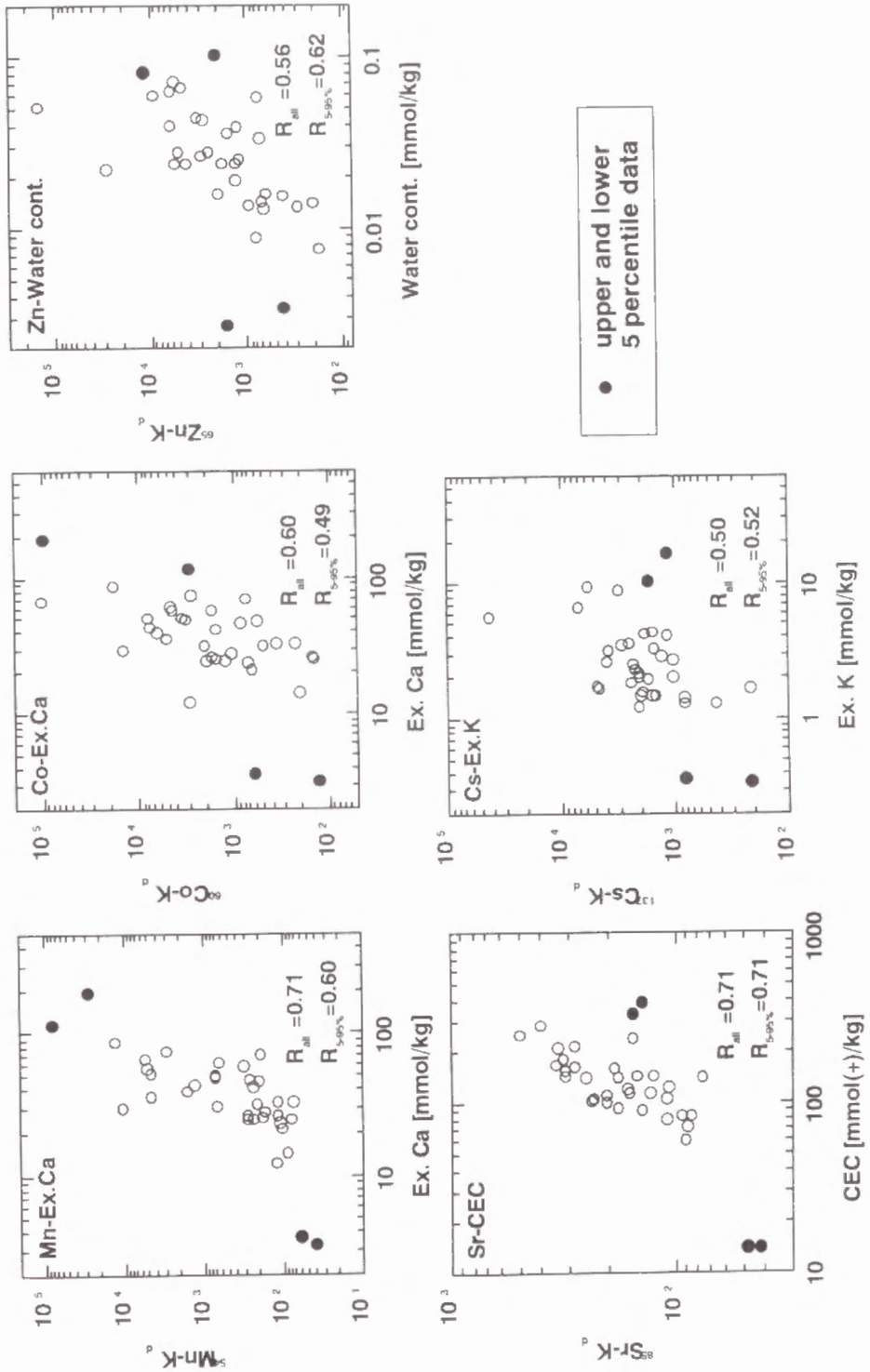

Figure 15. Plots of the K_ds versus the soil property having the highest correlation for each radionuclide; R in the figure is Pearson's correlation coefficient between their logarithms.

Table 12. Spearman's rank correlation coefficients between radionuclide K_ds and supernatant pH, EC and CEC/EC.

nuclide	pH	EC	CEC/EC
^{54}Mn	0.72	0.48	0.22
^{60}Co	0.69	0.39	0.23
^{65}Zn	0.28	0.01	0.32
^{85}Sr	0.08	-0.11	0.82
^{137}Cs	0.28	0.07	-0.03

3.3 DEVELOPMENT OF METHODS TO ESTIMATE K_d

3.3.1 Using electrical conductivity

Relationship between K_d and EC. As shown in the results of the experiments on effects of competing ions (section 3.2.2), it is clear that the K_d is affected by the competing ion concentration. Therefore, in this section, the relationships between the K_d and the supernatant EC are examined to discuss the possibility of using the EC for estimating the effect of competing ions. Plots of the K_d [mL g^{-1}] versus the supernatant EC [mS cm^{-1}] obtained in section 3.2.2 are shown in **Figure 16**. For all radionuclides, linear relationships are seen in the logarithmic graphs. The regression equations of the power function are

$$^{54}Mn: K_d=2.32\times10^{1}\times EC^{-0.75} \quad (25)$$

$$^{60}Co: K_d=1.83\times10^{2}\times EC^{-1.11} \quad (26)$$

$$^{65}Zn: K_d=8.57\times10^{2}\times EC^{-1.38} \quad (27)$$

$$^{85}Sr: K_d=1.87\times10^{1}\times EC^{-1.14} \quad (28)$$

$$^{137}Cs: K_d=1.75\times10^{2}\times EC^{-1.12} \quad (29)$$

Correlation coefficients between the observed and the predicted values are 0.99, 0.97, 0.93, 0.97, and 0.99 for ^{54}Mn, ^{60}Co, ^{65}Zn, ^{85}Sr, and ^{137}Cs, respectively. The index numbers of the above equations are less than zero and close to -1, which indicates that the amount of these radionuclides adsorbed onto soil would be almost inversely proportional to the soil solution EC.

Verification of the regression equations. The eqs. 25-29 were obtained under almost the same conditions, i.e. the same temperature, same soil-solution

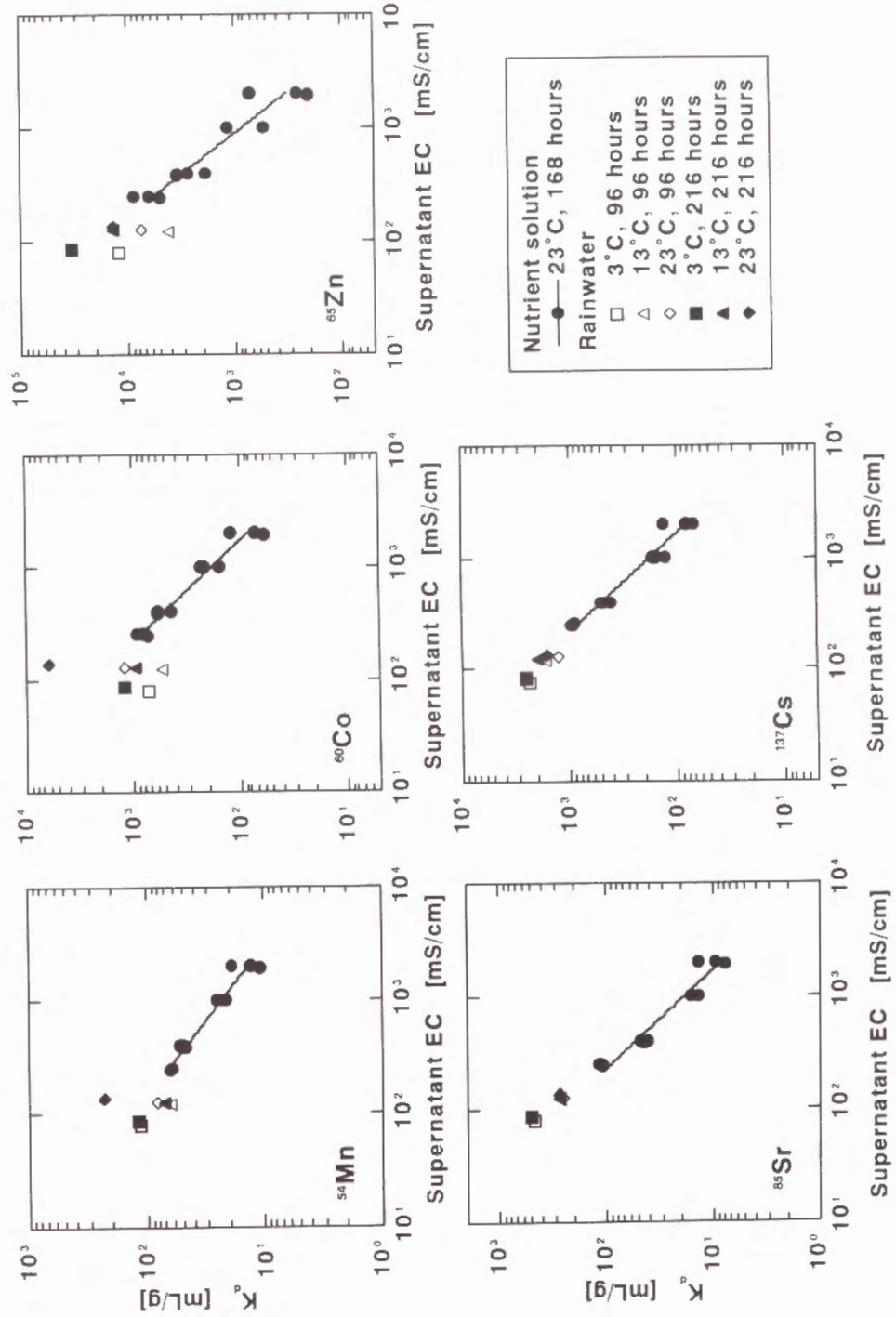

Figure 16. Plots of the radionuclide K_ds versus the supernatant EC; the solid lines in each graph are regression curves of the power function.

Table 13. The K_ds estimated from the supernatant EC using the regression equations shown in Figure 16 and the observed K_ds at 216 hours; the equations were obtained at 168 hours in another experiment.

radionuclide	temperature	estimated K_d [mL/g]	observed K_d [mL/g]	estimated/observed ratio
^{85}Sr	3 ℃	3.11×10^2	4.99×10^2	0.62
	13 ℃	2.03×10^2	2.79×10^2	0.73
	23 ℃	1.85×10^2	2.74×10^2	0.68
^{137}Cs	3 ℃	2.78×10^3	2.59×10^3	1.07
	13 ℃	1.82×10^3	1.99×10^3	0.91
	23 ℃	1.66×10^3	1.61×10^3	1.03

ratio, and same nutrient components (Table 8). Hence whether these equations are applicable to various cases is unclear. To verify their applicability under different conditions, the relationships between the K_ds and the supernatant EC obtained from the experiments on thermal effect (section 3.2.1) are also plotted in **Figure 16**. The K_ds estimated from the supernatant EC using the above regression equations are shown in **Table 13** for ^{85}Sr and ^{137}Cs with the observed K_ds obtained at 216 hours. The results of ^{54}Mn, ^{60}Co and ^{65}Zn are omitted in the table because no stable K_ds could be determined. The estimated K_ds of ^{85}Sr and ^{137}Cs agree well with the observed ones. Each error between the predicted value and the observed one is less than 38 % for ^{85}Sr and less than 9 % for ^{137}Cs; the accuracy of prediction is expected to increase if the regression equations were obtained for more diluted solution. These results suggest that reliable K_ds of Sr and Cs ions in the equilibrium state can be estimated from the soil solution EC. Because EC is easily and quickly measured, the method using EC for determining the K_d is considered to be very practical in actual fields. Particularly it would be an effective tool in a site where the salinity varies greatly in time and spatial scale or the salt accumulation is occuring.

Interpretation of the results. Generally, only salinity represented by EC is not sufficient information to determine the exact K_d; the ion composition in solution is also an important factor although the effect of competing ions is peculiar to each

radionuclide (Tanaka et al.,1991). We can assume that the majority of co-existing cations in a cultivated surface soil would be defined as calcium, magnesium, sodium, and potassium. The major exchangeable cation on the soil surface would be calcium (see Table 9). The contribution of other substances is considered to be relatively small because nutritional conditions must be adjusted for keeping crop growth. Each of these cations tends to form a hydrated ion having one or two positive electrical charge. High EC will be established when each cation in the solution has a high electric charge and a high mobility. The mobility of the cation is affected by the hydration situation. The ion hydrated by a smaller number of H_2O molecules tends to move faster, which contributes to higher EC. Also such a small ion has a greater affinity (higher selectivity) onto the soil surface and, as a result, prevents adsorption of radionuclide ions. This mechanism is schematically illustrated in **Figure 17**. The highly charged or little hydrated cation increases the solution EC and prevents sorption of co-existing radionuclides. Accordingly, the EC is considered to be more practical for the estimation of the K_ds, compared to each ion concentration, because it can evaluate the effect of ion composition changes. In addition, it has good advantages of being measured easily, quickly, and continuously.

3.3.2 Using cation distribution ratio (CDR) to estimate strontium K_d.

Materials and methods. To investigate the effects of solution properties, the strontium K_ds were collected by the same procedure in section 3.2.3 using riverwater obtained from the Naka River in Ibaraki, Japan. The chemical properties of the riverwater are shown in **Table 14**. The experiments were carried out for 10 soils (five upland soils : nos.1, 4, 6, 7, 10, and five paddy soils : nos.19, 21, 22, 23, 30) among 36 agricultural soils shown in **Table 15**. The results in section 3.2.3 are added to the following discussion.

Table 14. Chemical properties of the riverwater (contact solution).

pH	EC [mS/cm]	cation concentration [mmol/L]				
		Na^+	NH_4^+	K^+	Mg^{2+}	Ca^{2+}
7.2	0.175	0.810	0.006	0.076	0.271	0.640

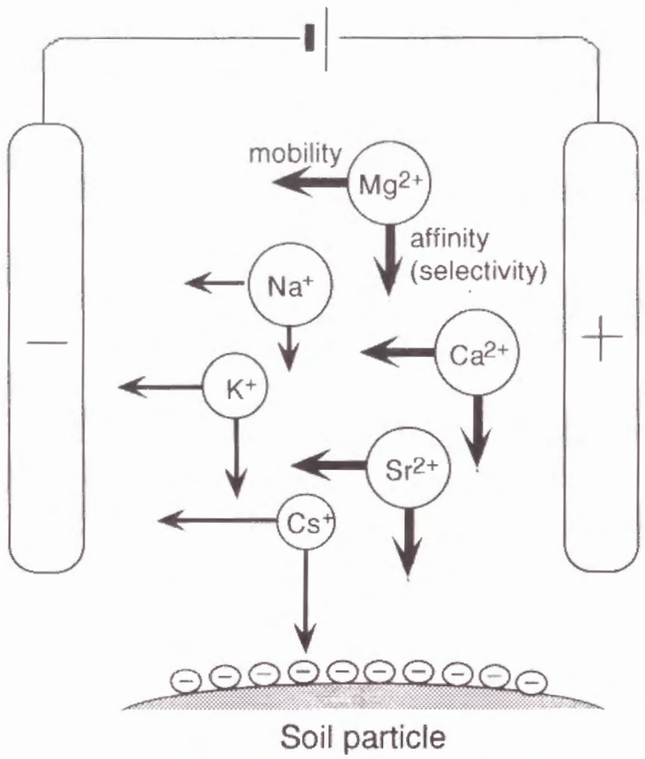

Figure 17. Illustration showing the mobility toward an electrode and the affinity (selectivity) onto soil surface of each ion.

Table 15. Chemical properties of the sample soils.

soil type	No.	CEC [mmol(+)/100g]	exchangeable K [mmol/100g]	exchangeable Ca [mmol/100g]	total Al [mmol/100g]	total C [g/100g]	cluster No.*
upland soil	1	11.4	0.52	2.78	334.3	0.73	1
	2	10.0	0.33	2.51	334.0	0.67	1
	3	11.4	0.87	3.91	255.0	1.68	1
	4	9.3	0.66	4.32	266.9	1.76	1
	5	13.9	1.33	11.56	315.8	2.50	1
	6	14.4	0.70	5.74	385.5	1.12	1
	7	25.4	0.39	7.51	407.7	6.06	3
	8	21.6	0.34	8.63	364.0	1.91	1
	9	11.9	0.85	2.60	304.7	1.03	1
	10	16.6	0.30	1.21	426.2	4.31	1
	11	14.2	2.00	3.60	356.9	2.54	1
	12	20.8	0.82	4.71	285.0	4.46	1
	13	1.4	0.07	0.33	240.5	0.07	1
	14	1.4	0.07	0.36	279.5	0.07	1
	15	10.4	1.13	2.96	283.7	1.07	1
	16	6.0	0.40	1.41	278.0	0.90	1
	17	14.4	1.75	3.10	441.8	1.26	1
	18	23.9	0.26	4.87	316.2	9.66	3
paddy soil	19	10.6	0.32	2.48	329.9	0.94	1
	20	10.3	0.37	2.46	295.4	0.74	1
	21	7.3	0.26	2.35	248.3	1.45	1
	22	16.1	0.41	6.67	349.9	2.45	1
	23	8.4	0.57	3.25	321.0	1.58	1
	24	8.4	0.54	3.25	316.5	1.44	1
	25	14.0	0.43	6.21	333.2	3.01	1
	26	8.0	0.25	2.64	300.2	0.48	1
	27	17.8	0.29	5.01	372.5	4.89	1
	28	15.3	0.46	4.19	348.4	2.05	1
	29	39.0	1.83	19.44	286.9	2.04	2
	30	14.0	0.73	5.78	322.5	1.72	1
	31	12.2	0.63	2.50	319.5	1.53	1
	32	33.5	0.33	7.03	364.7	10.21	3
	33	9.0	0.29	2.10	293.4	2.40	1
	34	11.0	0.51	3.14	298.0	1.46	1
	35	16.0	0.29	4.62	285.4	2.24	1
	36	28.6	3.27	5.14	344.0	2.00	4

* The cluster numbers were obtained from cluster analysis of the k-means method using the standardized data of soil properties; the samples have been grouped into four clusters.

Definition of cation distribution ratio (CDR). For the ten soils (nos. 1, 4, 6, 7, 10, 19, 21, 22, 23, 30), the concentrations of major cations (Na^+, K^+, NH_4^+, Mg^{2+}, Ca^{2+}) in the supernatant were measured by ion chromatography (Dionex, DX-300). The relationship between the total equivalent of these cations and the EC in the supernatant is shown in **Figure 18**. From the figure, the cation concentration are assumed to be proportional to the EC in the supernatant; the relationship is

$$\text{EC}\ [\text{mS cm}^{-1}] \approx 100 \times \text{Cation conc. in solution}\ [\text{mmol(+) mL}^{-1}] \quad (30)$$

Though radionuclide K_d can be theoretically calculated if the major competing ion is known (Wahlberg et al., 1965), it is difficult to clarify all the ion constituents in soil solution in an actual field. Eq.30 shows, however, that the effect of competing ions can be evaluated in a simple way using EC for agricultural soils although the composition of all ions including the anions affect the EC (Kamewada, 1991; Tanaka et al., 1991). In addition, from eq.30 and the definition of CEC, it is found that the ratio of CEC to EC (CEC/EC) indicates the total cation distribution ratio (0.01×CEC/Cation concentration) between the soil surface and soil solution; this value can have the same units as that of K_d [L kg^{-1}]. Here this ratio is termed a 'cation distribution ratio (CDR)'.

Correlation with strontium K_d. Correlations with the strontium K_ds and the soil properties are shown in **Figure 19.** Good correlations are found in relationships with CEC, Al, and C; these factors are considered to affect cation-exchange process. The plots of the strontium K_ds versus the CDRs are shown in **Figure 20**. The correlation with CDR is much higher (Spearman rank correlation coefficient =0.87) than those with other soil properties. The linear regression curve obtained by the least square method is

$$K_d = 2.1 \times \text{CDR} \quad (31)$$

From the above equation, the adsorption power of Sr is about twice that of the total cations. Using this simple relationship, we can easily get an accurate strontium K_d in the field by measuring only EC in the respective area. This method is considered to be much practical because the other soil properties such as soil pH and content of organic materials can be omitted in the measurement.

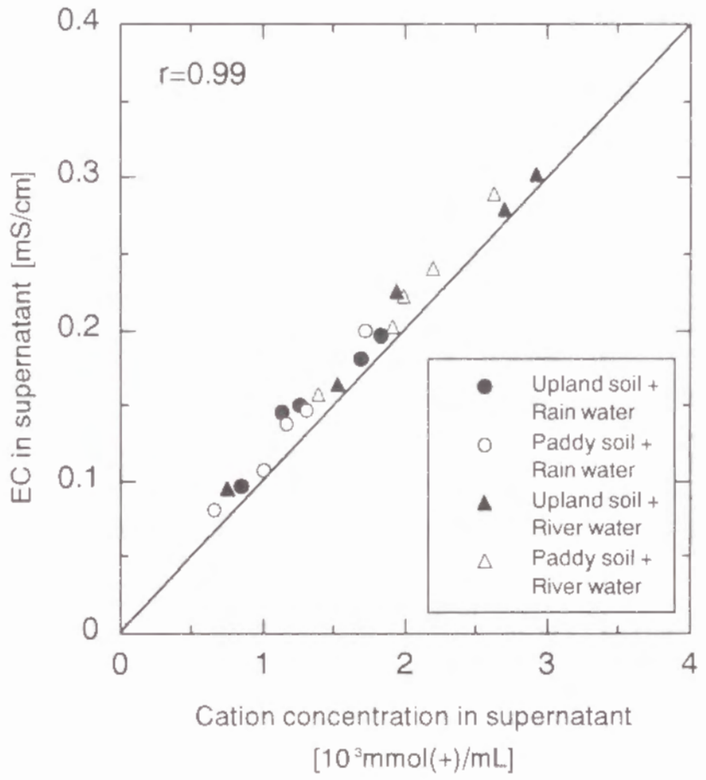

Figure 18. Relationship between the cation concentration and the EC in supernatants; the cation concentration was calculated by $Na^+ + K^+ + NH_4^+ + Mg^{2+} + Ca^{2+}$.

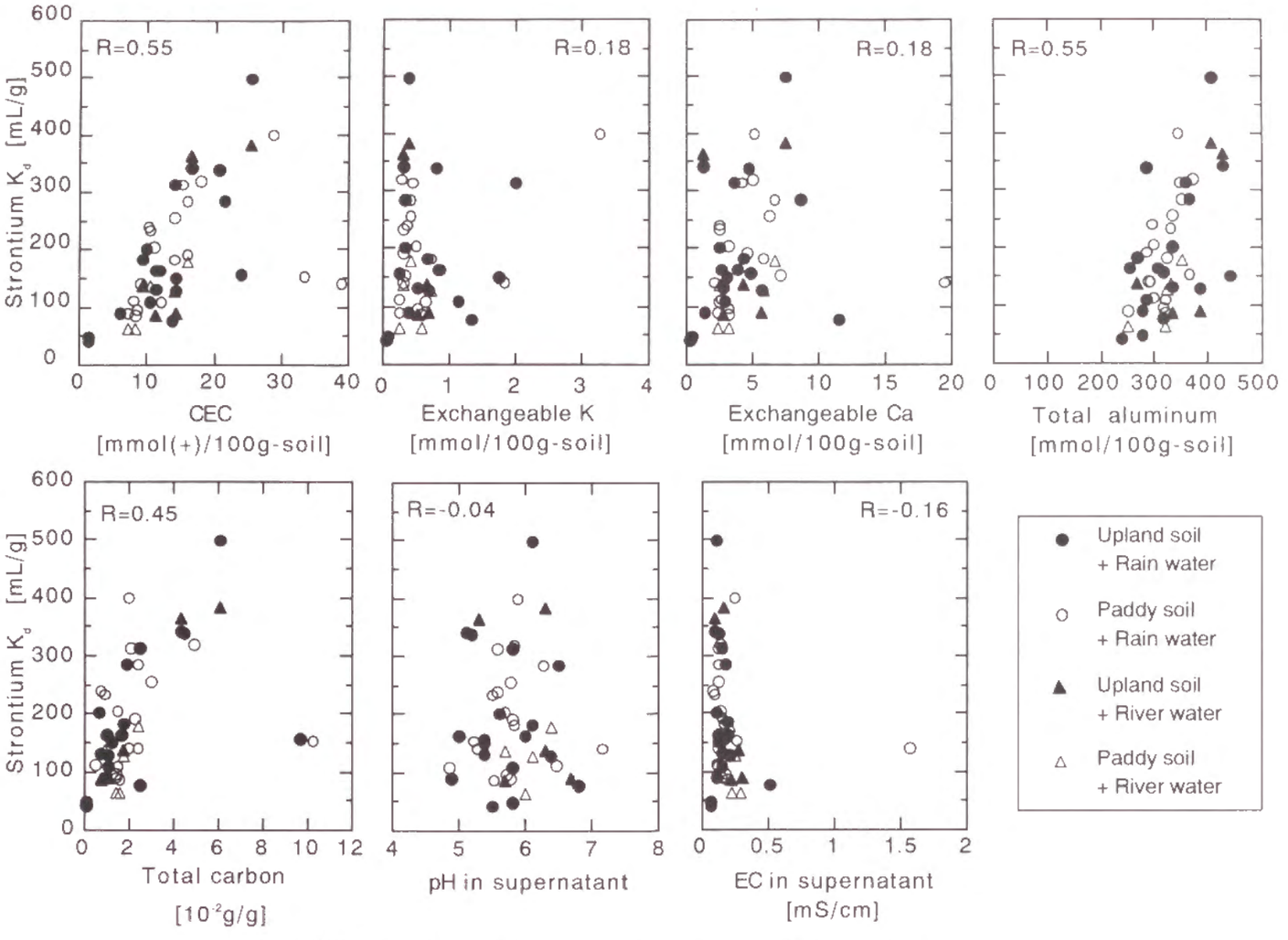

Figure 19. Relationships between strontium K_ds and soil or solution properties.

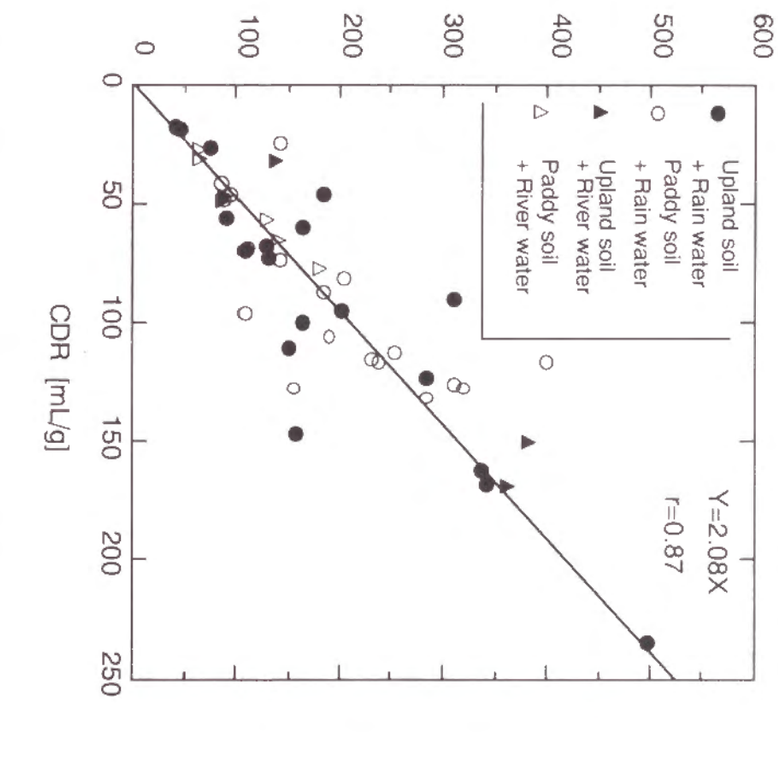

Figure 20. Plots of CDR values versus strontium K_ds; these are classified by experimental conditions.

Cluster analysis. Cluster analysis of the k-means method (Hartigan and Wong,1979) was carried out for the sampled soils using the standardized data of the soil properties shown in **Table 15** (Water cont., CEC, ex.K, ex.Ca, total Al, and C); these properties are considered to relate to ion exchange processes. The soils were grouped into four clusters. The cluster number obtained for each soil is also shown in **Table 15**. Most soils belong to cluster 1 (31 soils); these soils can be assumed to be 'general agricultural soil'. The sample in cluster 2 (1 soil) has a high amount of exchangeable Ca. Cluster 3 (3 soils) includes high total carbon samples. The sample in cluster 4 (1 soil) has a high amount of exchangeable K. The clustered plot of the relationship between K_d and CDR is shown in **Figure 21**. Though the linear regression curve for cluster 1 samples is almost the same as eq.31, the correlation coefficient obtained from only cluster 1 samples is higher (r=0.90) than that obtained from all soil samples (r=0.87). Accordingly, this regression equation is considered to be suitable for general agricultural soils.

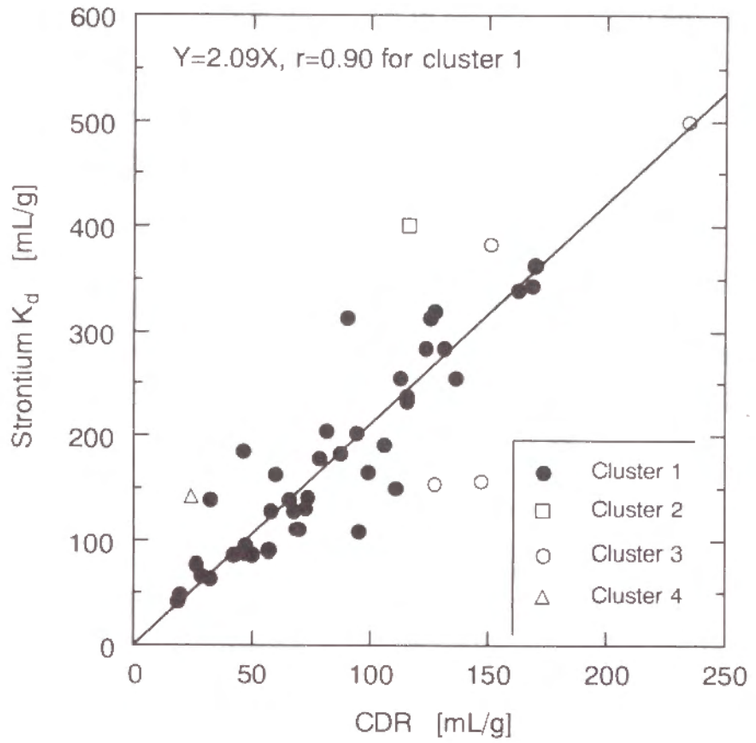

Figure 21. Plots of CDR values versus strontium K_ds; these are grouped by the cluster number obtained from cluster analysis of the k-means method.

CHAPTER 4. ACCURACY IMPROVEMENT OF SOIL-TO-PLANT TRANSFER FACTOR

4.1 INTRODUCTION

As mentioned in section 2.4.2, many T_fs have already been collected and default T_fs have been given for some crops (see Tables 2-4). However, large variabilities have been found in the reported T_fs (Ng et al., 1982; Ng and Hoffman, 1983; Keen and Frissel, 1983; Yasuda and Uchida, 1993c), and the cause of the T_f variation is unclear at present. Though some margin for safety should be added to the T_f to prevent underestimation of the internal exposure dose, suitability of those margins can not be judged unless the variational characteristics of the T_f have been clarified.

The variational characteristics of the T_fs have been statistically investigated according to the classification variables (Ng and Hoffman, 1983; Frissel and Koster, 1988; Yasuda and Uchida, 1994). It is still difficult, however, to improve the reliability of the T_fs by such a qualitative manner. Although the quantitative information on the growth condtions is required, the available data are lacking because most of the T_fs have been obtained empirically without giving attention to the surroundings. We need more efforts to clarify the primary factors affecting T_fs and to quantify their effects.

4.2 VARIABILITY OF REFERENCE T_fS

If sufficient T_fs have been obtained in a certain field, further suitable T_fs could be determined on the basis of their statistical characteristics, e.g., their frequency, distribution type, mean value, standard deviation, and percentile values. In this section, the T_fs of Sr (T_f-Sr) and the T_fs of Cs (T_f-Cs) reported by IUR (1987) were statistically analyzed. The IUR report has stated many T_fs on a dry weight basis (Bq g^{-1} dried vegetation per Bq g^{-1} dried soil) with some information such as crop name, crop part, soil type, soil-pH, and measurement methods (field/ lysimeter/ pot). Three leafy plants: cabbage, spinach, and grass were analyzed here; clovers were included as grass. In the analysis, the T_fs obtained for non-edible parts (whole or roots) and those in peculiar soils such as clay, silt, and gravel, except loam and sand, were eliminated.

4.2.1. Probability distributions

The probability distributions of T_f-Sr and T_f-Cs are shown in **Figures 22 and 23**, respectively. The p value is the probability obtained by the Shapiro-Wilk test (Shapiro and Wilk, 1965) when assuming the T_fs of each nuclide had normal or log-normal distributions. In all cases, the T_fs show log-normal type distributions with a significance level of 1%.

The numbers (n), the geometric mean (GM), the standard deviation of the common logarithm (SDCL), and the percentile values (10, 50, and 90 %) of the T_fs are shown in **Table 16** for T_f-Sr and **Table 17** for T_f-Cs for each crop. For all crops, the GMs of T_f-Sr (4.1 for cabbage, 2.4 for spinach, and 1.7 for grass) are larger than 1, while those of T_f-Cs are lower (0.063 for cabbage, 0.41 for spinach, and 0.19 for grass). From the GMs, it is considered that T_f-Cs have a larger variability than T_f-Sr because the former vary by a factor of 2.9 and the latter by 6.5. The SDCLs tend to be larger for T_f-Cs (0.59 for cabbage, 0.32 for spinach, and 0.72 for grass) than for T_f-Sr (0.35 for cabbage, 0.18 for spinach, and 0.31 for grass). The results of the t-test between T_f-Cs and T_f-Sr, using their logarithms, show that there is a significant difference ($p<0.01$) for each crop.

4.2.2. Comparison among crops

The GMs are ordered differently among the crops; the descending order of T_f-Sr is cabbage, spinach, and grass, and that of T_f-Cs is spinach, grass, and cabbage. The results of the t-test between the crops (cabbage-spinach, spinach-grass, and grass-cabbage), using the logarithms of T_fs, show that there is a significant difference ($p<0.01$) for each nuclide. Accordingly, it is recommended that a specific and appropriate T_f should be adapted for each crop although one default T_f tends to be given in general radiological assessments (USNRC, 1977; IAEA, 1982).

4.2.3. Effect of soil type

To investigate the effect of soil properties, the T_fs were analyzed according to soil type classification: loam and sand. The probability distributions for each crop are shown in **Figure 24** for T_f-Sr and **Figure 25** for T_f-Cs. The statistical values (n, GM, SDCL, and 10, 50, and 90 percentile values) are stated for each soil type also in **Tables 16 and 17**. In all cases, the GM is larger for sand than

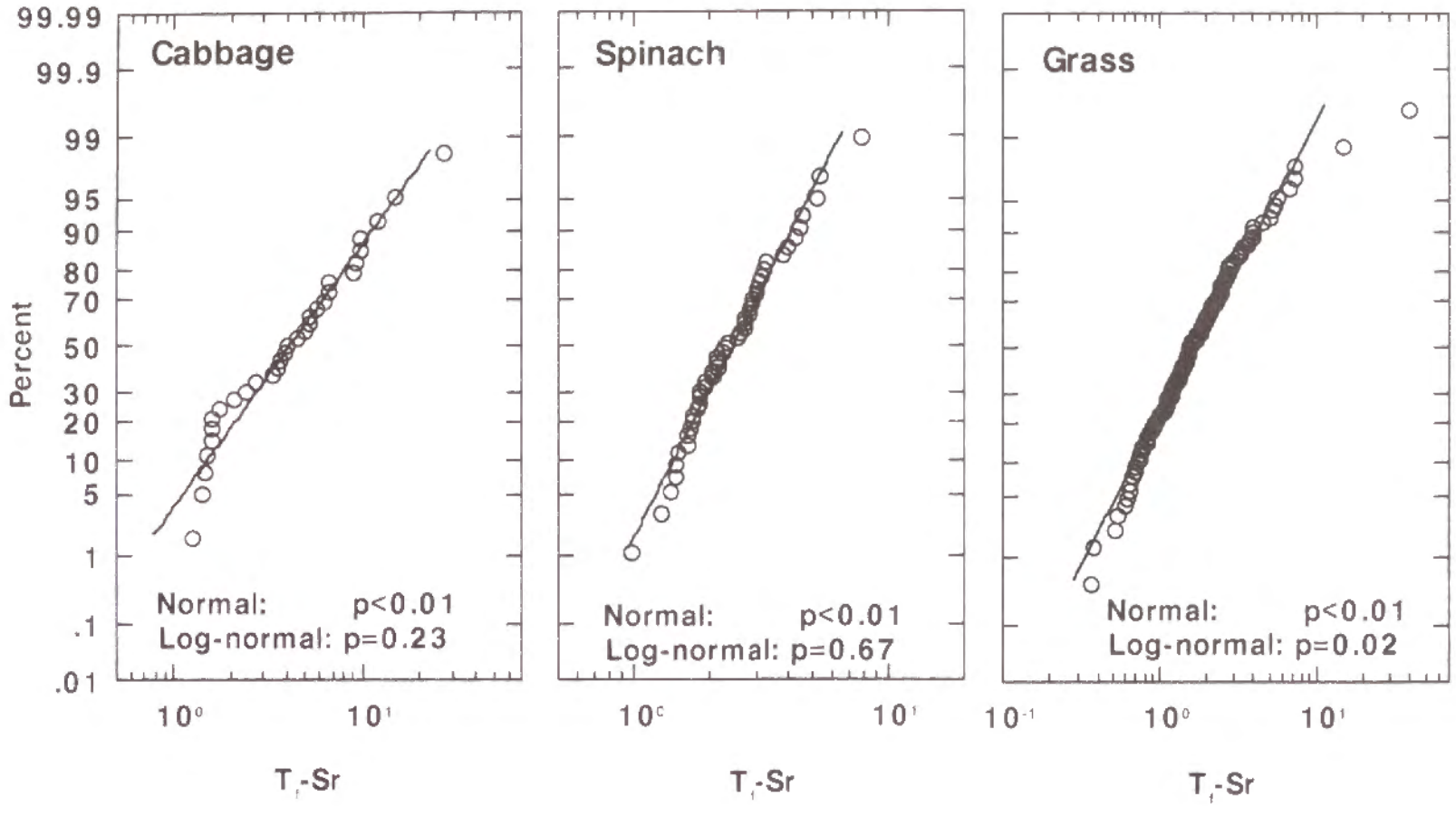

Figure 22. Probability distributions of strontium T_fs in the IUR report for three leafy plants; p value is the probability obtained in the Shapiro-Wilk test for each distribution type.

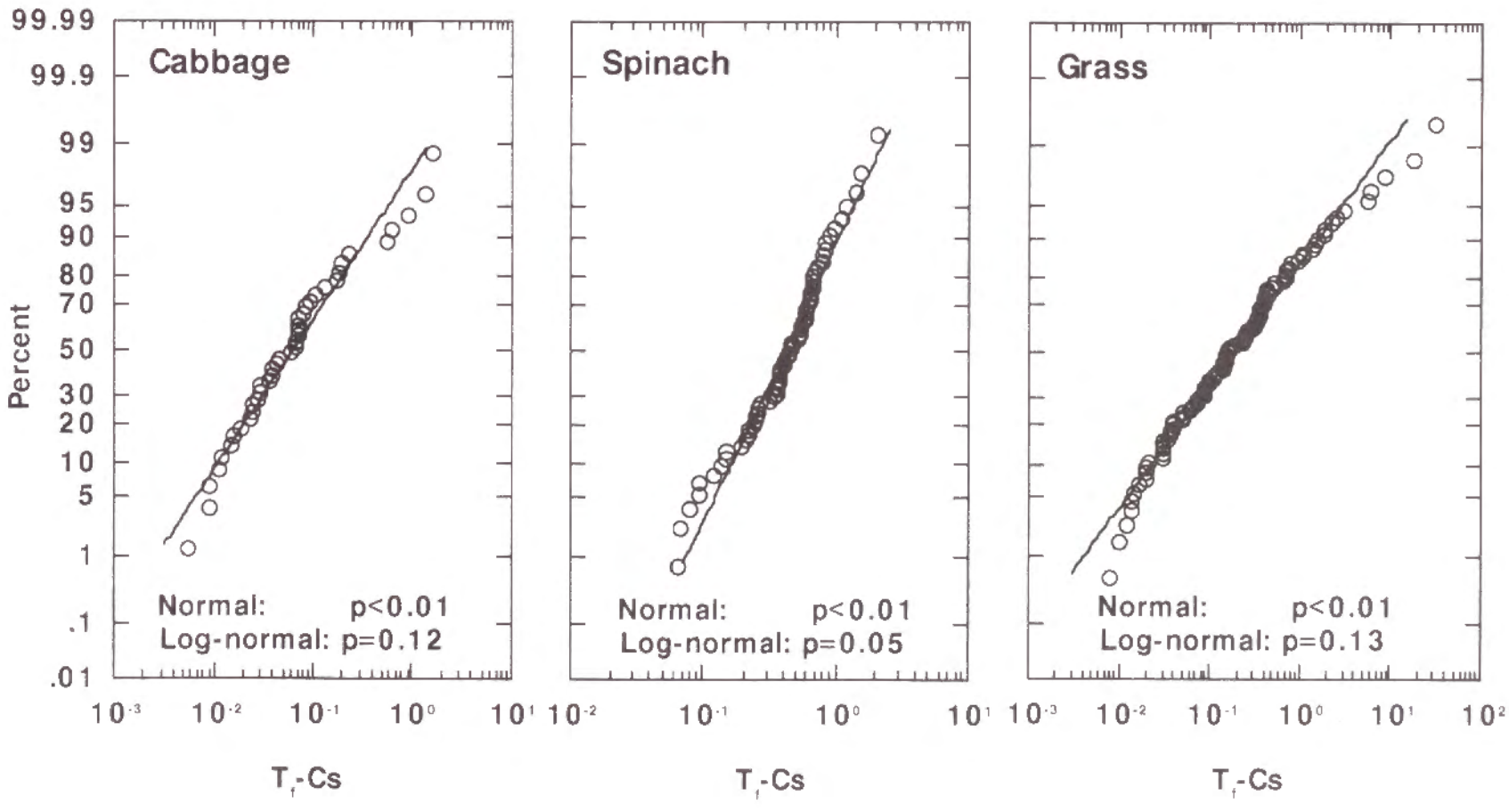

Figure 23. Probability distributions of cesium T_fs in the IUR report for three leafy plants; p value is the probability obtained in the Shapiro-Wilk test for each distribution type.

Table 16. Statistical characteristics of strontium T_fs on a dry weight basis.

crop	soil	n	geometric mean	std. dev. com. log.	percentile value		
					10%	50%	90%
cabbage	loam+sand	31	4.10	0.347	1.46	4.05	11.56
	loam	17	3.77	0.289	1.50	4.55	8.30
	sand	24	4.55	0.414	1.36	3.87	19.40
spinach	loam+sand	48	2.43	0.184	1.45	2.31	4.54
	loam	28	2.08	0.130	1.44	2.00	3.16
	sand	20	3.00	0.210	1.79	2.96	5.39
grass	loam+sand	116	1.70	0.312	0.72	1.55	4.00
	loam	53	1.28	0.285	0.61	1.26	2.64
	sand	63	2.15	0.297	0.90	2.06	5.13

Table 17. Statistical characteristics of cesium T_fs on a dry weight basis.

crop	soil	n	geometric mean	std. dev. com. log.	percentile value		
					10%	50%	90%
cabbage	loam+sand	40	0.063	0.594	0.012	0.064	0.624
	loam	23	0.031	0.218	0.009	0.028	0.154
	sand	17	0.166	0.294	0.040	0.093	1.386
spinach	loam+sand	68	0.409	0.320	0.138	0.455	0.917
	loam	40	0.342	0.333	0.093	0.395	0.725
	sand	28	0.527	0.269	0.215	0.560	1.141
grass	loam+sand	102	0.191	0.722	0.021	0.160	1.789
	loam	34	0.158	0.473	0.025	0.168	0.588
	sand	68	0.210	0.818	0.020	0.150	2.562

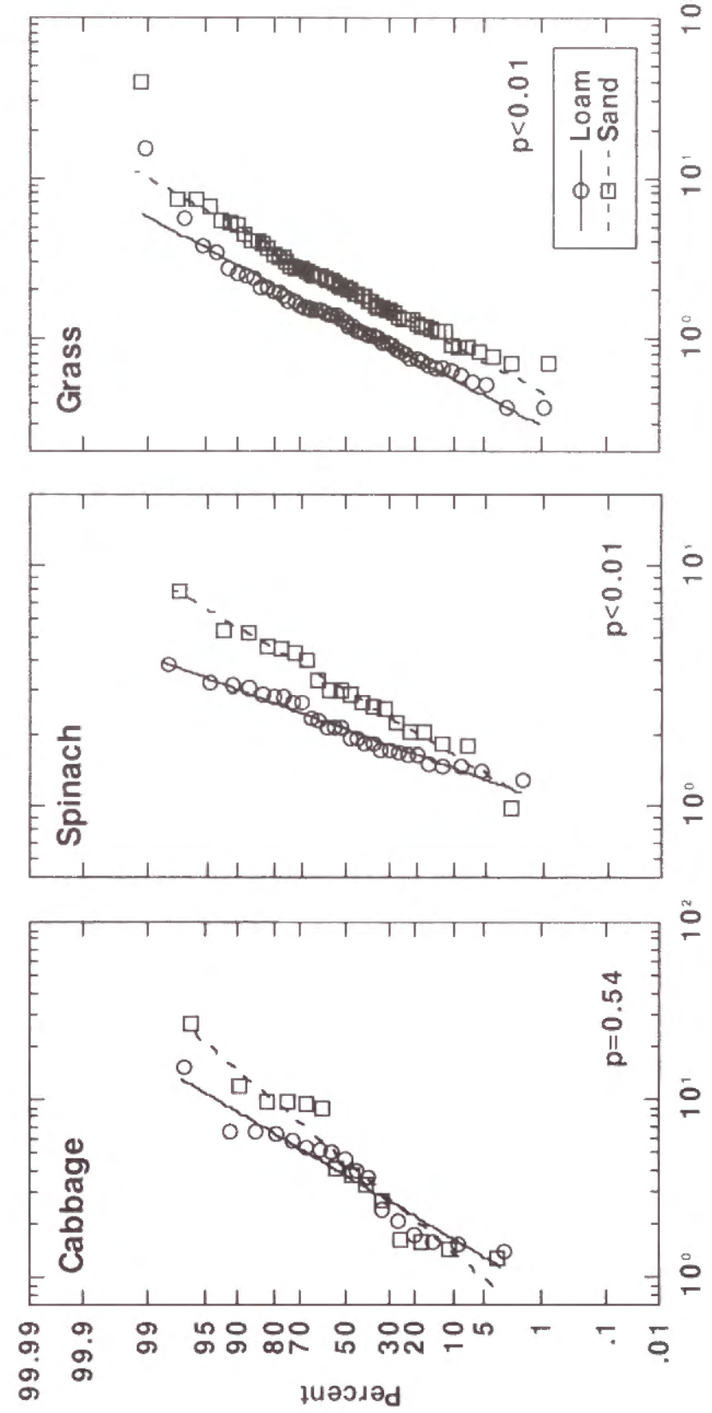

Figure 24. Probability distributions of strontium T_fs in the IUR report according to loam and sand soil types. The p value is the probability obtained in the t-test between loam and sand using the logarithms of the T_fs.

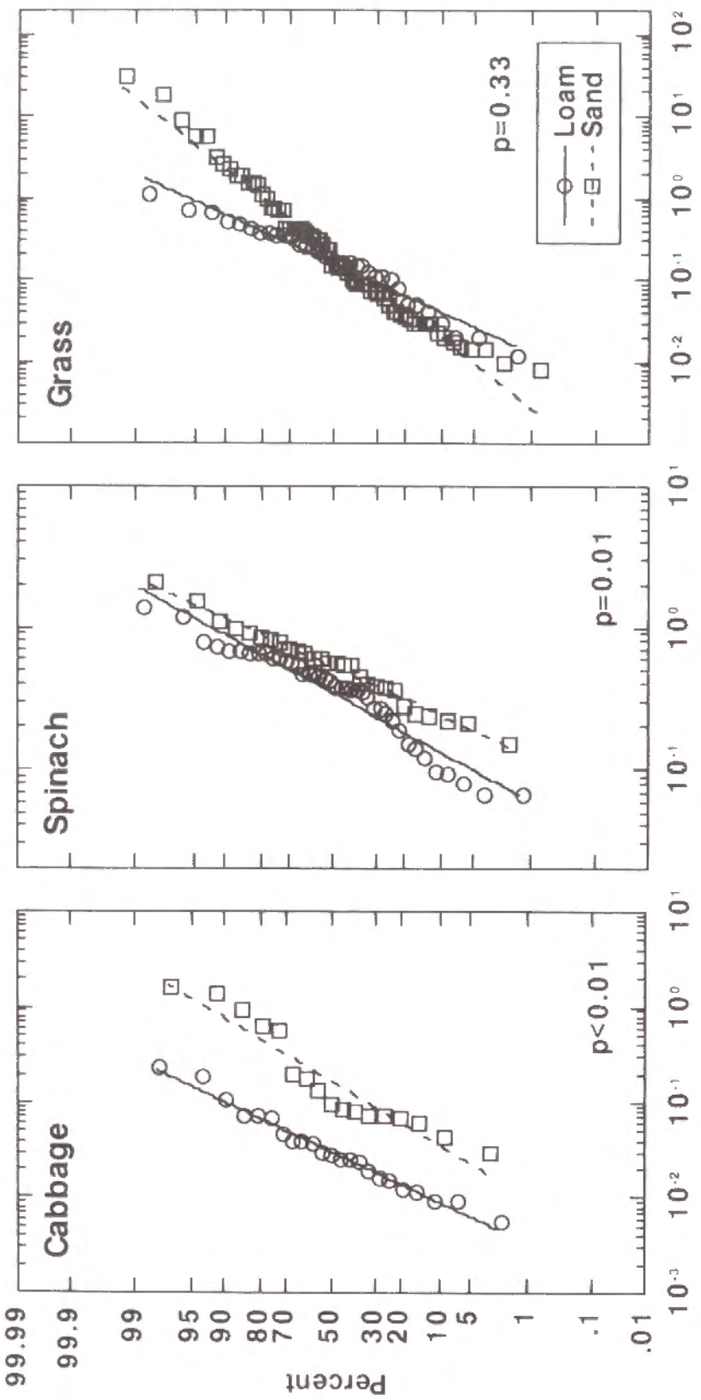

Figure 25. Probability distributions of cesium T_fs in the IUR report according to loam and sand soil types. The p value is the probability obtained in the t-test between loam and sand using the logarithms of T_fs.

that for loam. In the results of the t-test using the logarithms of the T_fs, a significant difference ($p<0.05$) between loam and sand is found in four cases: T_f-Sr of spinach and grass, and T_f-Cs of cabbage and spinach. These results are considered due to the difference of solubility of each nuclide between the two soil types; the solubility decreases with the adsorption capacity of soil. Sand is considered to have a lower adsorption capacity than loam. The SDCLs are also larger in sand except for one case: T_f-Cs of spinach. It has already been reported that T_f variation becomes smaller with increase of particle size of soil (Ng and Hoffman, 1983). These results can be explained according to the kinetics of solute uptake by crop roots as follows.

- Due to increased solute concentration caused by the solubility increase, a linear relationship between the solute uptake rate by roots and the solute concentration in soil will not be established (Epstein and Hagen, 1952; Shaw et al., 1992).
- When the adsorption capacity of the soil is small, the solute concentration at the root surface will greatly change followed by a large variation of the solute uptake by roots (Yasuda and Uchida, 1993a).

For loamy soil, classification of soil type is considered to be effective to reduce uncertainty of T_f because the SDCL for loam tends to be smaller than that for all samples (loam+sand). When the plots of the T_fs appear to be parallel between loam and sand in the figures -- in case of T_f-Sr for grass and T_f-Cs for cabbage --, the uncertainty of T_f is expected to be reduced also for sand based on soil type classification.

4.2.4. Suitability of default T_f

For major radionuclides, default T_fs are stated in some reports (see **Table 4**). In the IAEA report (IAEA, 1982), the default values of T_f -Sr and T_f -Cs for forage plants on a dry weight basis are 2 and 0.1, respectively, and it is noted that site-specific investigations are recommended for sandy soils.

In this section, the suitability of these default T_fs were examined in comparison with the grass T_fs. **Table 18** shows the ratio of the default T_f to the geometric mean of the grass T_fs and the percent of the default T_f assuming it obeys the probability distribution of the grass T_fs. The default T_f-Sr is found to be very close to the geometric mean of grass T_f-Sr for loam+sand and for sand while it has a rela-

Table 18. Comparison of the default T_fs for forage plant with the grass T_fs.

soil type	Sr (T_f=2)		Cs (T_f=0.1)	
	ratio to the geometric mean	percent[a]	ratio to the geometric mean	percent[a]
loam+sand	1.18	0.59	0.52	0.35
loam	1.56	0.76	0.63	0.34
sand	0.93	0.46	0.48	0.35

[a] cumulative percent of the same value with the default T_f in the probability distribution of grass T_fs.

tively large error for loam. Hence the default T_f-Sr is considered to be suitable for sandy soil. The default T_f-Cs is about half of the geometric mean of grass T_f-Cs for all cases and the errors are almost the same. This suggests that the default T_f-Cs in the IAEA report have a large potential to underestimate the transfer from soil to grass.

4.3 NUMERICAL STUDIES ON UNCERTAINTY OF T_f

The results in section 4.2 suggested that the variation of T_fs attributes partially to the difference of soil type. Here numerical simulations were carried out in accordance with the kinetics of solute uptake by plants roots to confirm mechanistically effects of soil types (loam/sand) on the T_f variation. In the calculation, different K_d values taken from the literature were given to each soil type.

4.3.1 Models and conditions

Plant roots can be assumed to be cylindrical tubes (Nye and Tinker, 1977; Barber, 1984). According to this assumption, the material movement in soil around the roots can be approximated by the following radial-flow equation:

$$\frac{1}{r}\frac{\partial}{\partial r}\left(rD\frac{\partial \theta C_l}{\partial r}\right) - v\frac{\partial \theta C_l}{\partial r} = \frac{\partial}{\partial t}C_l(\theta + \rho_b K_d) \qquad (32)$$

where r is the distance from the root center [cm]; D, the dispersion coefficient [$cm^2\ min^{-1}$]; θ, the volumetric water content in soil [$mL\ cm^{-3}$]; C_l, the material concentration in soil solution [$mg\ mL^{-1}$]; v, the water velocity [$cm\ min^{-1}$]; t, the time [min]; ρ_b, the bulk soil density [$g\ cm^{-3}$]; and K_d, the distribution coefficient

0.933. For silt, the mean of the common logarithm is 3.91 and the standard deviation of the common logarithm, 0.423. Also, to consider changes of meteorological conditions, the water uptake rate (Q) was given as a randomized value. Though the probability distribution of Q is unknown, the normal distribution with the mean value of 0.1 [$mL\ min^{-1}$] and the standard deviation of 0.03 were given in this simulation.

Eq.34 and eqs.37-39 were differentiated by the Crank-Nicolson method. The program was written in FORTRAN and performed 300 times for each soil type on a workstation (SUN).

4.3.2 Results and discussion

The calculated solute concentrations near the root surface under some conditions are shown in **Figure 26**. Though the material concentrations at the root surface are increasing in all cases, the K_d value affects the increase rate. When the K_d is smaller, the solute concentration at the root surface varies on a larger scale. The increase of Q also affects the solute concentration at the root surface; its effect is considered to become larger in smaller K_d.

The probability distribution of T_{fc} calculated for each soil type is shown in **Fig-**

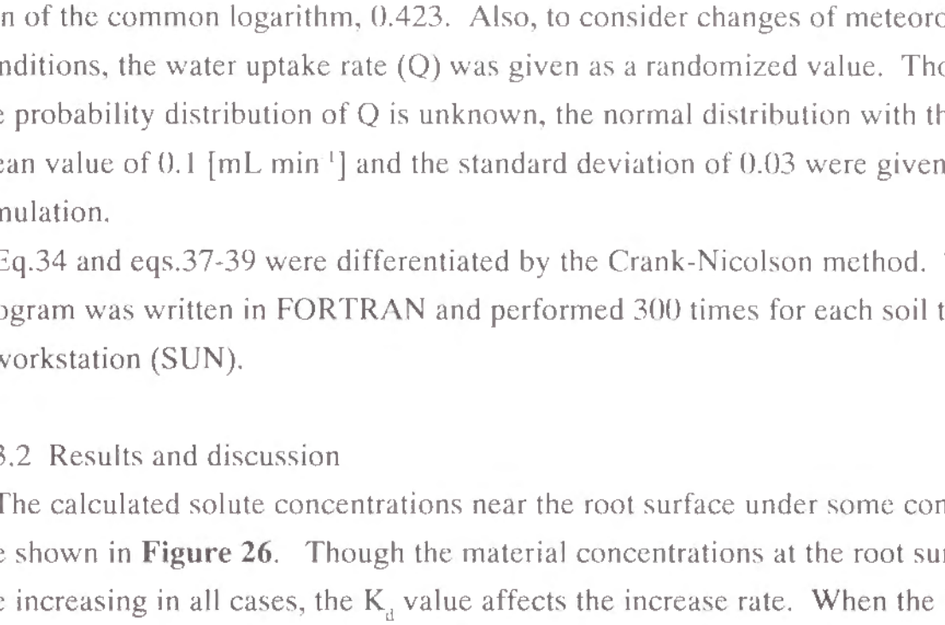

Figure 26. Calculated concentrations in soil solution near the root surface at 6 days for 6 cases: two K_d values × three Q values.

To validate the simulation results in comparison with the observed T_f, the transfer factor in the period of 6 days (T_{fc}) is defined as follows:

$$T_{fc} = \frac{f_e \cdot \int_{t'=0}^{t'=6d} 2\pi r_o l \cdot a_p \cdot C_l(r_o, t')dt'}{C_{soil}} \qquad (40)$$

where f_e is the concentration coefficient into edible parts by their weight [g^{-1}]; $C_l(r_o,t')$, the solute concentration at $r=r_o$ and $t=t'$ [$\mu g\ mL^{-1}$]; and C_{soil}, the concentration in soil by soil weight [$\mu g\ g^{-1}$]. The T_{fc} is the ratio of the material amount concentrated into the plant edible parts to the $C_{soil.}$ The C_{soil} is calculated by

$$C_{soil} = C_b \cdot (\frac{\theta}{\rho_b} + K_d) \qquad (41)$$

Parameters used in the simulation are summarized in **Table 19**. The K_d was given as a randomized value of the log-normal distribution because K_ds for agricultural soils show a log-normal type distribution as shown in Figure 14. The statistical data of the cesium K_ds obtained in two soil types, sand and silt, were adapted from the literature (Sheppard et al., 1984). For sand, the mean of the common logarithm is 2.67 and the standard deviation of the common logarithm is

Table 19. Parameter values used for the numerical simulation of root uptake; the K_ds were given as randomized values of the log-normal distribution and the Qs, those of the normal distribution.

parameter*		value
θ	[mL/cm^3]	0.20
ρ_b	[g/cm^3]	1.6
L	[cm]	100
r_d	[cm]	10
r_o	[cm]	0.10
D	[cm^2/min]	6.0×10^{-4}
a_p	[$mL/cm^2/min$]	1.0×10^{-3}
f_e	[1/g]	0.1
C_b	[$\mu g/mL$]	1.0

[$mL\ g^{-1}$]. Here we assume that the moisture conditions are constant and the soil properties are homogeneous. The water velocity v can be expressed by

$$v = -\frac{Q}{2\pi L\theta} \cdot \frac{1}{r} \qquad (33)$$

where Q is water uptake rate by a whole plant [$mL\ min^{-1}$]; and L, the total root length [cm]. Then eq.32 becomes

$$D\frac{\partial^2 C_l}{\partial r^2} + \frac{D+q}{r}\frac{\partial C_l}{\partial r} = R_f \frac{\partial C_l}{\partial t} \qquad (34)$$

where

$$R_f = 1 + \frac{\rho_b K_d}{\theta} = \text{const.} \qquad (35)$$

$$q = \frac{Q}{2\pi L\theta} \qquad (36)$$

In the initial state, the material is assumed to be uniformly distributed in soil. That is,

$$\text{I.C.} \qquad C_l = C_b \qquad \text{at} \quad t = 0 \qquad (37)$$

The material is assumed to be supplied at the outer boundary where the initial concentration is maintained at a distance of $r=r_d$:

$$\text{B.C.1} \qquad C_l = C_b \qquad \text{at} \quad r = r_d \qquad (38)$$

According to previous studies (Epstein and Hagen 1952; Epstein, 1966; Barber, 1984), the root uptake kinetics are described by the Michaelis-Menten type equation and the solute influx to plant roots is independent of water absorption. Here it is assumed that the solute concentration is much lower than the Michaelis constant, i.e., the material is assumed to be taken up by roots in proportion to its concentration in the solution at the root surface; the proportionality coefficient is referred to as a root absorption coefficient. Hence the inner boundary condition at the root surface can be given by

$$\text{B.C.2} \qquad D\frac{\partial \theta C_l}{\partial r} - v\theta C_l = a_p C_l + \frac{\partial}{\partial t} C_l(\theta + \rho_b K_d) \qquad \text{at} \quad r = r_o \qquad (39)$$

where a_p is the root absorption coefficient [$mL\ cm^{-2}\ min^{-1}$]; and r_o, root radius [cm].

ure 27. Both the distributions are log-normal. In comparison between the two soil types, the geometric mean of T_{fc} is larger for sand than for silt. The variation (the standard deviation of the common logarithm) is also larger for sand. In the correlation analysis, an adequate correlation is obtained between the K_d and the T_{fc}; Spearman's rank correlation coefficient is -1.0 for each soil type. This means that the difference of T_{fc} distribution between the two soil types can be explained by the difference of K_d distribution. While no correlation is obtained between Q and T_{fc}, the absolute values of Spearman's rank correlation coefficients are less than 0.1 for both soil types. Then changes of water uptake rate are considered to have a relatively small effect on T_f under the conditions in this simulation. These results suggest that the uncertainty of T_f depends on the variability of K_d to a large extent.

Since loam is defined as a soil having a relatively high content of silt (USDA, 1975), these simulation results agree qualitatively with the observed T_f distributions shown in **Figures 24 and 25**, in which the variation of T_f tends to be larger for sand than for loam. It is expected that more reliable T_fs are estimated on the basis of the K_ds in each cultivated soil.

Though the potential variation of other parameters and their effect on T_f need to be discussed more in future studies, the results obtained in this study suggest that the K_d variability in space or time scale is of primary importantance in evaluating the uncertainty of T_f, as is migration in soil. Practical methods to determine the suitable K_d, like the CDR method proposed in section 3.3.2, should be developed. The accurate K_ds obtained by using such methods in real fields will greatly improve the reliability of the model prediction in a terrestrial environment.

4.4 PROPOSAL OF A NEW MODEL TO ESTIMATE T_f

The statistical approach such as that shown in section 4.2 needs many observed data under carefully designed conditions because many factors are at work in a complex manner in actual fields; it is difficult to evaluate the quantitative effect of each factor. An effective alternative to the statistical approach is to develop a model which is able to predict the T_f using physical (quantitative) parameters corresponding to the growth conditions. Such a model enables us to determine suitable T_fs for various cases having different conditions. The model, however,

should be simple enough to allow practical use.

In this section, a new model is proposed to estimate T_f in accordance with the kinetics of solute uptake by plant roots. It is validated for strontium in pot experiments.

4.4.1. Model derivation

A schematic diagram of the internal structure of a plant root is shown in **Figure 28**. Major ions in the soil solution enter the free space (apoplasm) in the cortex and reach the xylem through the symplasm (Nye and Tinker, 1977). Here it is assumed that the radionuclide ions rapidly disperse in the solution and are absorbed into the free space while keeping the following equilibrium relationship:

$$C_f = K_{fl} \cdot C_l \qquad (42)$$

where C_f is the concentration in the free space [Bq mL^{-1}]; K_{fl}, the distribution ratio of the radionuclide ions between the free space and the soil solution [-]; C_l, the concentration in the soil solution [Bq mL^{-1}]. The C_l in an equilibrium state can be given by

$$C_l = \frac{C_{soil}}{\theta_l + K_d} \qquad (43)$$

where C_{soil} is the radionuclide concentration in soil [Bq g^{-1}]; θ_l, soil water content by weight [mL g^{-1}]; K_d, the soil-solution distribution coefficient [mL g^{-1}] which is considered to represent sorption intensity of soil. Because the K_ds for cationic nuclides such as strontium are much larger than θ_l (Sheppard et al., 1984), eq.43 is approximated as

$$C_l \approx \frac{C_{soil}}{K_d} \qquad (44)$$

The radionuclide ions in the free space must pass through the symplasm to reach to the xylem (Nye and Tinker, 1977), but this process is not clearly understood. The radionuclide ions are assumed to pass the symplasm through the surrounding semipermeable membranes in accordance with the electrochemical potential gradient. Therefore, when the potential gradient is constant throughout the roots, the concentration in the xylem (C_x) [Bq mL^{-1}] can be calculated by

$$C_x = P_{xf} \times C_f \qquad (45)$$

where P_{xf} is the concentration ratio between the xylem and the free space [-] which

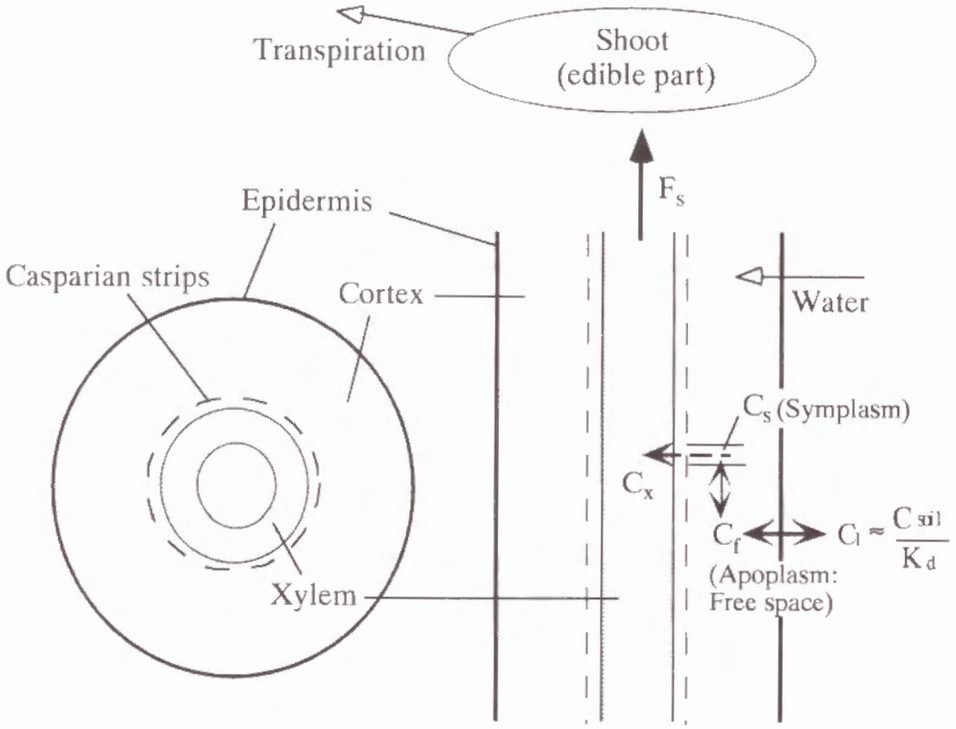

Figure 28. Schematic illustration of root uptake kinetics.

will be affected by the electrochemical condition. The radionuclide ions in the xylem are considered to be transported to the shoot passively by solution movement (Nye and Tinker, 1977). Hence the flux of the radionuclide ion to the shoot (F_s) [Bq h^{-1}] can be given by

$$F_s(t) = w(t) \times C_x \qquad (46)$$

where w(t) is the water flux to the xylem [mL h^{-1}] which can be assumed to be equal to the transpiration rate because the volume of water transpired is much larger than those retained in the plant tissue. If volatilization from the shoot and relocation to the root can be neglected, the radionuclide concentration in the shoot at the end of growth period (C_{crop}) [Bq g^{-1}] is given by

$$C_{crop} \approx \frac{\int_0^T F_s(t)dt}{w_e} \qquad (47)$$

where T is the growth period [h]; w_e, the shoot weight [g]. Substituting eqs.42-46 into eq.47 gives

$$C_{crop} = \frac{\int_0^T w(t) \times P_{xf} \times K_{fl} \times C_{soil}\ dt}{w_e \times K_d} \qquad (48)$$

When the P_{xf} and K_{fl} can be assumed to be constant during the growth period, eq.48 becomes

$$C_{crop} = \frac{W}{w_e} \times \frac{P_{xf} \times K_{fl}}{K_d} \times C_{soil} \qquad (49)$$

where W is total transpiration volume during the growth period [mL]. Here two, intensive parameters are defined as follows:

$$S_c = P_{xf} \times K_{fl} \qquad (50)$$

$$T_c = \frac{W}{w_e} \qquad (51)$$

Then eq.49 is simplified to

$$C_{crop} = \frac{S_c \times T_c}{K_d} \cdot C_{soil} \qquad (52)$$

This equation indicates that the soil-to-plant transfer factor, T_f, is described by the following equation:

$$T_f = \frac{S_c \times T_c}{K_d} \qquad (53)$$

The S_c is termed here as the selective absorption coefficient [-] which is considered to be a characteristic value for each plant species and T_c, the transpiration coefficient [mL g^{-1}] which is assumed to be related to nutritional and meteorological conditions. The S_c is not the same as the selectivity coefficient (Dorp et al., 1979) which does not include translocation to the shoot. Although many radionuclide K_ds have been obtained (Sheppard et al., 1984), those for agricultural soils are limited and they have shown large variations (RWMC, 1990). It seems difficult to determine an *in situ* K_d except for strontium which can be estimated from the CDR (section 3.3.2). In the following sections, the strontium T_f was estimated by eq. 53 for a leafy vegetable and examined for its accuracy by comparing it with the result in a pot experiment.

4.4.2. Materials and methods

Measurement of selectivity coefficient. The S_c was measured in solution-culture experiments using a pot filled with nutrient solution including about 10 kBq ^{85}Sr (half life: 65 d) in a carrier-free state. A leafy vegetable, *Brassica campestris* (13 days old) was used as a sample plant. The initial weight of each plant is shown in **Table 20**. To examine effects of competing ions on the S_c, five solutions having different ion concentrations were used; they were standard nutrient solution (pH, 5.3; EC, 2.3; NH_4^+, 1.7 mmol L^{-1}; K^+, 7.8 mmol L^{-1}; Mg^{2+}, 1.9 mmol L^{-1}; Ca^{2+}, 3.6 mmol L^{-1}) and four solutions diluted from the standard one

Table 20. The conditions and the results in the 70 hour solution-culture experiments.

solution					crop (*Brassica campestris*)			
dilution factor	initial EC [mS/cm]	initial pH	final EC [mS/cm]	final pH	initial whole weight [g]	final weight		transpiration [mL]
						whole [g]	shoot [g]	
1	2.30	5.3	2.28	5.5	1.07	2.24	1.51	19.1
2	1.19	5.2	1.11	5.6	1.15	2.23	1.49	98.9
3	0.78	5.2	0.73	5.8	1.38	2.85	1.73	28.1
5	0.49	5.1	0.43	5.6	0.69	1.35	0.89	70.3
10	0.27	4.9	0.19	5.4	0.63	1.47	0.98	43.1

by factors of 2, 3, 5, and 10, respectively. The chemical properties of each solution are also shown in Table 20. The total ion concentration in the solution was represented by the EC because a linear relationship has been established between the total cation concentration and solution EC for agricultural soils (Yasuda and Uchida, 1993b). The pots were set in a growth chamber (Rheem, Series 1500) which was kept at 23 °C; they received about 25,000 lx for 12 hours per day. The water surface was covered with styrofoam to restrict evaporation. The weight of each pot was measured to get the transpiration volume during the experimental period. At 70 hours after setting the pots in the chamber, the weight of each fresh shoot, and EC and pH in the solution were quickly measured. Each fresh shoot was then put into a plastic vial and analyzed for γ-ray intensity with a Ge-detector (Ortec GEM-30185) coupled to a multichannel analyzer (Seiko, EG&G 7800).

Soil-culture experiments. To validate eq.53, soil-culture experiments were carried out. Soils were collected from five upland fields in Japan; they were dried at room temperature for about one month, and sieved to 2 mm. Their chemical properties are shown in **Table 21**. The cation exchange capacity (CEC), exchangeable K and exchangeable Ca were measured by Schollenberger's method (Shollenberger and Simon, 1945). For the Al determination, the sample was decomposed in a strong acid solution (60 % HNO_3 15mL+48 % H_2SO_4 4mL+46 % HF 5mL) (Geological Survey of Japan, 1980). Potassium was determined with an atomic absorption spectrometer (Shimadzu, AA646) and Ca and Al, with an inductively coupled plasma spectrometer (ICP-AES; Seiko, SPS1100H). Carbon was analyzed with a reference-type thermal conductivity detector (Yanagimoto,

Table 21. Chemical properties of soils; each soil was collected from an upland field in Japan.

run no.	CEC [mmol(+)/100g]	exchangeable K [mmol/kg]	exchangeable Ca [mmol/kg]	total Al [mmol/kg]	total C [g/kg]
1	21.6	4.1	120.8	3640	19.1
2	34.2	12.5	119.5	4122	98.9
3	13.2	14.6	60.4	3751	28.1
4	34.2	12.5	80.8	3703	70.3
5	16.6	3.6	16.9	4262	43.1

CHN corder MT-3) (Dojoyoubun-bunsekihou-iinkai, 1983).

A seed of *Brassica campestris* was planted in each plastic pot containing 150g of soil mixed with about 100 kBq ^{85}Sr. Some fertilizer (KNO_3: 80 mmol+ $NH_4H_2SO_4$: 14 mmol) and deionized water (75 mL) were also added to each pot. The pots were set in the growth chamber (Rheem, Series 1500) which was kept at 23 °C and received about 25,000 lx for 12 hours per day. The soil surface was covered by an aluminum sheet to restrict evaporation of soil solution. The experimental periods were 15 days for four samples and 20 days for one sample which did not grow well. At the end of the respective periods, the weights of each pot and each fresh shoot were quickly measured. The fresh shoot was put into a plastic vial and analyzed for γ-ray intensity. The pH(H_2O) of the soil was measured with the soil:solution ratio of 1:2.5 (Jpn. Soc. Soil. Sci. & Plant Nutr., 1988). The EC in the soil solution was estimated from the EC in the supernatant by the following equation:

$$EC = \left(1 + \frac{W_{water}}{\theta_i \times W_{soil}}\right) \cdot EC_m \qquad (54)$$

where W_{water} is the volume of pure water added before measuring EC [mL]; W_{soil}, the soil weight [g]; and EC_m, the EC in the supernatant [mS cm^{-1}].

4.4.3. Results and discussion

The S_c [-] was obtained by dividing the uptake amount of ^{85}Sr [Bq] by both the transpiration volume [mL] and the ^{85}Sr concentration in the solution [Bq mL^{-1}]. The relationship between the S_c and the solution EC is shown in **Figure 29**. A linear relationship is found between the logarithms of the S_c and the EC; the S_c is inversely proportional to the EC. The regression equation is

$$S_c = 1.3 \times (EC)^{-0.72} \qquad (55)$$

where the units of EC are [mS cm^{-1}]. This result suggests that the salinity (the osmotic potential) in the solution affects the strontium transfer into the roots.

It has been presented that the strontium K_d for agricultural soils can be estimated reliably from the value of soil CEC divided by solution EC (section 3.3.2). The regression equation is

$$K_d = \frac{2 \times CEC}{EC} \qquad (56)$$

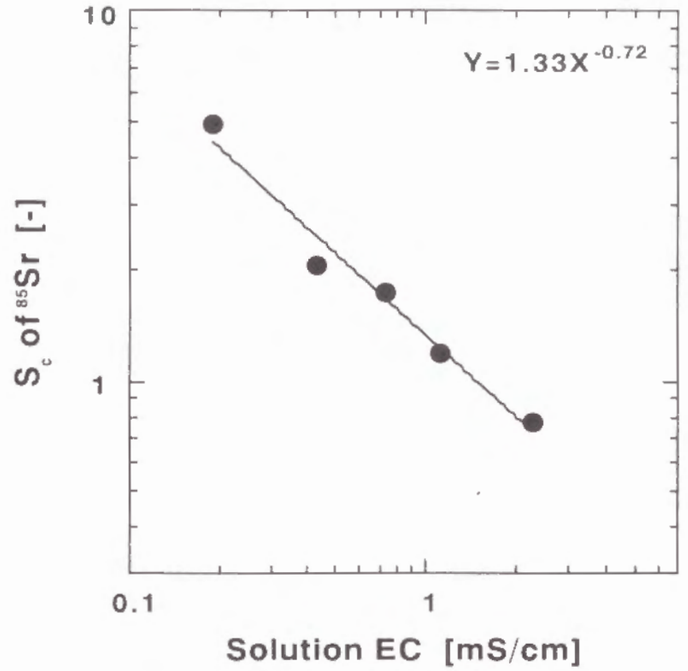

Figure 29. Plot of solution EC versus selective absorption coefficient (S_c) for *Brassica campestris*.

where CEC is the cation exchange capacity of the soil [mmol(+) $100g^{-1}$].

By substituting eqs.55 and 56 into eq.57, T_f is described as

$$T_f = \frac{0.63 \times (EC)^{0.28} \times T_c}{CEC} \qquad (57)$$

This equation means that the strontium T_f for *Brassica campestris* can be estimated from three environmental values, i.e., solution EC, soil CEC, and transpiration coefficient (T_c). In other words, strontium T_f has a potential to vary greatly due to changes of these factors. The K_d and T_f calculated by eqs.56 and 57 are plotted in **Figure 30** as a function of soil solution EC.

The S_c, K_d and T_f estimated from the actual EC by eqs.55, 56, and 57, respectively, are shown in **Table 22**. The experimental conditions differ to a large extent among the samples; the shoot weight varies by a factor of 5.5, the soil-pH(H_2O) by 1.5, and the estimated EC by 3.0. The parameters also differ; the estimated T_c varies by a factor of 3.9; the S_c, by 2.2; the K_d, by 1.5; and the T_f, by 5.8. Regardless of these differences, the T_fs calculated by eq.57 agree well with the observed ones; the ratio between calculated and observed values is 0.8-1.3.

Table 22. Experimental and estimated values and comparison of estimated T_fs with observed ones.

soil no.	growth period [day]	shoot weight at harvest [g]	T_c [mL/g]	soil-pH (H_2O) [a]	EC in soil solution [b] [mS/cm]	estimated K_d [L/kg]	estimated S [-]	estimated T_f [-]	observed T_f [-]	ratio of est./obs.
1	15	1.34	32.1	6.5	1.15	39.4	1.20	0.98	0.83	1.18
2	15	0.71	26.8	5.7	1.62	44.3	0.94	0.57	0.68	0.84
3	15	0.89	27.0	7.3	0.79	35.1	1.58	1.20	0.91	1.32
4	15	3.02	18.9	5.5	2.37	30.3	0.71	0.45	0.50	0.89
5	20	0.55	74.6	4.8	1.05	33.2	1.29	2.88	2.92	0.99

[a] $pH(H_2O)$ was measured by the batch technique with soil-water ratio of 1: 2.5.

[b] the EC in the soil solution was calculated by eq. .

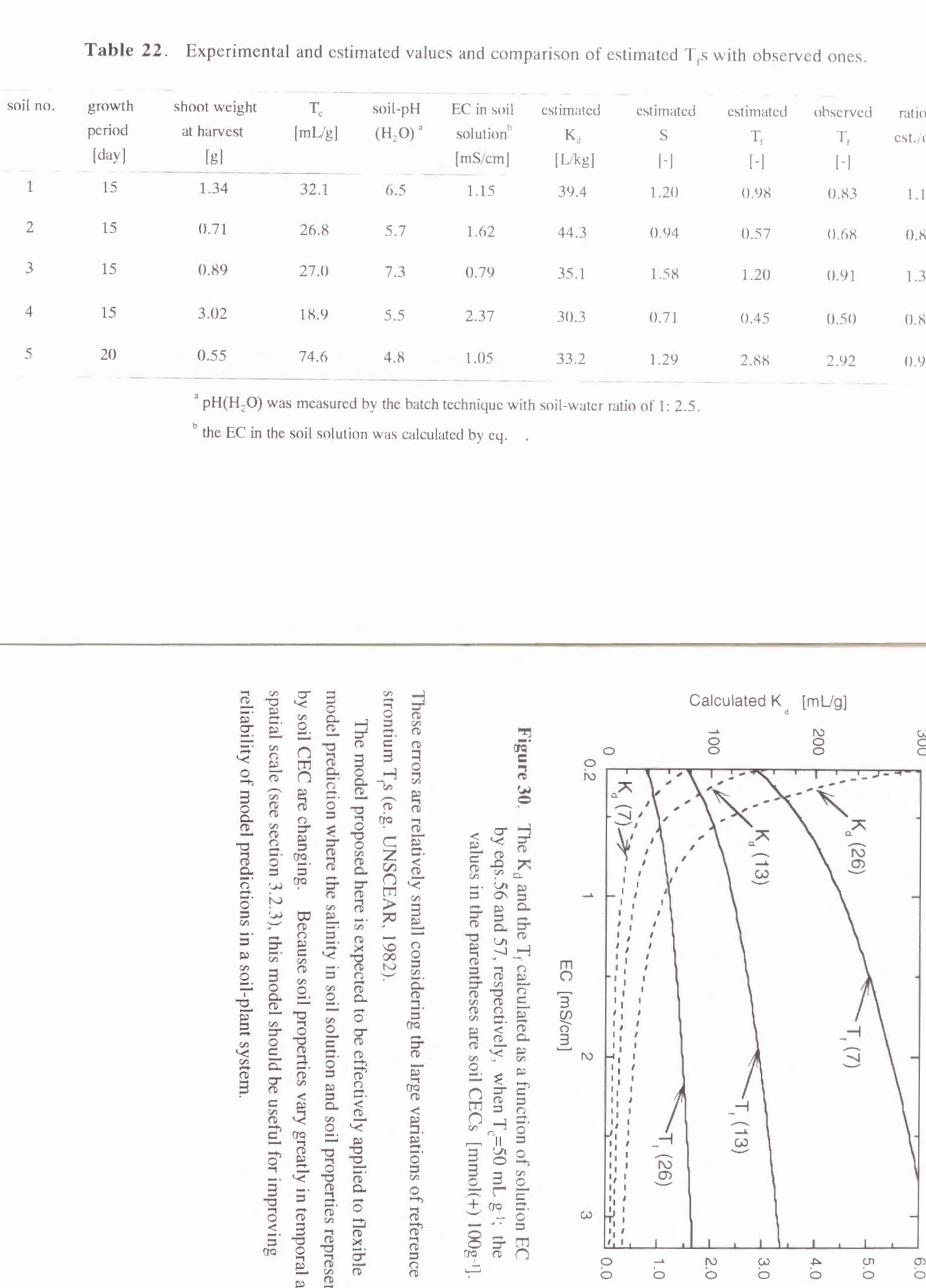

Figure 30. The K_d and the T_f calculated as a function of solution EC by eqs.56 and 57, respectively, when T_c=50 mL g^{-1}; the values in the parentheses are soil CECs [mmol(+) $100g^{-1}$].

These errors are relatively small considering the large variations of reference strontium T_fs (e.g. UNSCEAR, 1982).

The model proposed here is expected to be effectively applied to flexible model prediction where the salinity in soil solution and soil properties represented by soil CEC are changing. Because soil properties vary greatly in temporal and spatial scale (see section 3.2.3), this model should be useful for improving reliability of model predictions in a soil-plant system.

CHAPTER 5. RELIABILITY IMPROVEMENT IN ENVIRONMENTAL IMPACT ASSESSMENTS

5.1 INTRODUCTION

The ultimate goal of environmental impact assessments is to develop a relationship between the source term of a pollutant and the resulting risks. When cultivated land gets highly contaminated, we must judge the safety of the agricultural products and, if necessary, restrict the cultivation to prevent ingestion of the pollutants. In general, the level of contamination in agricultural soil is evaluated by the concentration of each pollutant in the soil. This concept is based on transfer of materials from soil to crop edible parts -- followed by transfer to man -- being proportional to their concentration in the soil. The proportionality coefficient is referred to as a soil-to-plant transfer factor (T_f). To decrease the adverse health effects caused by ingesting the contaminated crops, the T_f used for the evaluation should be large enough not to be below the actual T_f. As a result, many soils should be judged as 'contaminated' and would be restricted for the agricultural activities. Such a countermeasure, however, will induce serious social problems for the farmers (IAEA, 1989a), e.g., loss of land, necessary food, and income. In the determination of the derived intervention level (DIL), we must consider these social and economic problems in addition to health effects upon consumers. The establishment of appropriate DIL needs a reliable model prediction having reliable parameters being suitable to the field under consideration. When sufficient data on the parameters have been obtained there, the suitable parameter values can be determined based on the frequency of the observations. In cases without sufficient data, default values in references as shown in Tables 3-5 will be applied. However, reference parameters such as K_d and T_f would cause some errors because of their variabilities (Ng et al., 1982; Ng and Hoffman, 1983; Keen and Frissel, 1983; Frissel and Koster, 1988; Yasuda et al., 1993c) which have been difficult to be incorporated.

This chapter presents new, reliable techniques for environmental impact assessments by utilizing the results already obtained in this dessertation. The statistical information shown in Chapters 2 and 3 is applied to decision-making on surface soil cleanup and to determination of cultivation intervention level. The

flexible model developed in section 4.4 is also applied to risk analysis of contaminated agricultural land and its usefulness is discussed. It is expected that the framework of the process presented here will be applicable to other chemical contaminants. However, the suitability of the assessment process shown here as an example needs to be re-evaluated continually with updated information such as environmental conditions, numbers of concerned people, and cost-benefit situations.

5.2 DICISION MAKING FOR SURFACE SOIL CLEANUP

Surface soil contaminated by hazardous materials has a potential to adversely affect residents' health. Derived intervention levels (DILs) on selected countermeasures such as evacuation and relocation have been presented for the purpose of protecting the residents' health (e.g. IAEA, 1986b; ICRP, 1993). However, clear criteria are not yet available on whether the soil should be cleaned up or not. Cleanup is an important countermeasure for protecting residents from not only external irradiation, but also social and economic damages due to loss of their land.

On the basis of social situations, the people concerned with soil contamination can be categorized into three major groups: residents, responsible parties, and cleanup workers. It is considered that each group has a different interest and each will decide whether the soil should be cleaned up or not from a different viewpoint. When facing soil contamination, they should ask three different questions. The first question will arise from residents who want to know

"Is it safe to live there ? "

Risk analysists must answer this question based on the predicted health risk to the residents. If the soil is judged to be unsafe, a decision on proper countermeasures should be made as soon as possible not to harm the residents. In this step, responsible parties will ask the question:

"Which countermeasure is best ? "

Cost-benefit consideration is a primary subject for the people who are responsible for financing the countermeasures. Cleanup is not necessarily justified by them if it costs more than other countermeasures. When cleanup is chosen after cost-benefit consideration, the last question will be asked by the workers involved in the activities. This question is

"Are the cleanup activities safe ? "

The safety level in this stage should be able to protect workers' health which is as important as that of the residents.

This section offers principles necessary for answering the above questions, i.e. making decisions on safety of residence, advantages of the countermeasures, and safety of cleanup activities. As an example, the DILs of ^{137}Cs are calculated based on the principles for cleanup of contaminated surface soil. Cs-137 is the radionuclide which has brought about the most serious problems to residents of Russia and Europe since the Chernobyl accident.

5.2.1. Decision on safety of residence

The safety level for residence is defined as the concentration below which a resident will maintain a normal life expectancy. The life expectancy is assumed to be guaranteed when they do not exceed the dose limit which has been derived from the data of radiation effect on human health. Then the principle for deriving safety level for residence is defined as follows:

The dose that each resident will get in his lifetime should be less than the dose limit.

To calculate the dose that residents will get, changes of the radionuclide concentration in soil must be estimated for the expected period of their lives. The radionuclide concentration in surface soil can be assumed to change exponentially with time (e.g. NCRP 1984) as follows:

$$C_{soil}(t) = C_{soil}(0) \cdot e^{-\lambda \cdot t} \qquad (58)$$

where $C_{soil}(t)$ is the radionuclide concentration in the surface soil at time=t [Bq kg^{-1}]; and λ, the effective decay constant [y^{-1}].

Assuming that the concentration declines by radioactive decay and downward migration into deeper soil, λ can be expressed as

$$\lambda = \lambda_d + \lambda_i \qquad (59)$$

where λ_d is the radioactive decay constant [y^{-1}]; and λ_i, the rate of removal due to downward migration [y^{-1}]. Each term can be described in detail by using more specific parameters as follows:

$$\lambda = \frac{\ln 2}{T_H} + \frac{v_w}{W_{soil} \times (\theta_l + K_d)} \qquad (60)$$

where T_H is the half-life of the radionuclide [y]; v_w, the downward water velocity [L m^{-2} y^{-1}]; W_{soil}, the surface soil density [kg m^{-2}]; θ_l, the soil water content by weight [L kg^{-1}]; and K_d, the soil-solution distribution coefficient [L kg^{-1}]. The K_d is obtained as the ratio between the concentration in soil solid and that in liquid phase.

The cumulative dose that each resident will get additionally in his life (D_{life} [Sv]) can be estimated as follows:

$$D_{life} = \int_0^{T_{life}} F_{conv} \times C_{soil}(0) \times e^{-\lambda \cdot t} dt \qquad (61)$$

where T_{life} is the life expectancy [y]; and F_{conv}, the conversion coefficient from radionuclide concentration in the surface soil to the dose equivalent per resident [Sv yr^{-1} per Bq kg^{-1}]. Equation 61 can be transformed to

$$D_{life} = \frac{C_{soil}(0)}{\lambda}(1 - e^{-\lambda \cdot T_{life}}) \qquad (62)$$

The general values of the parameters included in eqs. 58-62 are shown in **Table 23**. It should be noted, however, that these values have uncertainties. For example, the K_d, an empirical parameter, varies greatly depending on physical, chemical, and biological conditions that are constantly changing in the actual fields. When evaluating D_{life} of each resident, we need to consider these uncertainties which accompany it.

Table 23. Parameters and the values used in the decision-making for surface soil cleanup.

parameter	definition	value*
T_h	half-life by radioactive decay	30 [y]
v_w	downward water velocity	740 [L m^{-2} y^{-1}]
W_{soil}	surface soil density	240 [kg m^{-2}]
θ_l	soil water content by weight	0.14 [L kg^{-1}]
T_{life}	life expectancy	30 [y]
F_{conv}	conversion coeff. from radionuclide conc. in surface soil to dose equivalent per resident	3.96×10^{-6} [Sv yr^{-1} per Bq kg^{-1}]
$N_{resident}$	number of residents	100 [man]
m_{comp}	annual compensation cost per resident	50,000 [$ man^{-1} y^{-1}]
A	residential area	10^6 [m^2]
N_{worker}	number of workers involved in cleanup	10 [man]

* The values of the v_w and the θ_l are taken from Hoffman and Baes (1979); the W_{soil}, USNRC (1977); and the F_{conv}, USDOE (1988).

Figure 31 shows a probability distribution of Cs-K_ds obtained for agricultural soils collected throughout Japan. It is found that they have a log-normal type distribution rather than a normal type. From the results in the Shapiro-Wilk test, shown as p values in the figure, the Cs-K_d distribution is judged to be log-normal type. Then, in this study, the Cs-K_ds are given as probabilistic values that have the log-normal distribution in Figure 31; i.e. the geometric mean is 1.85×10^3 L kg^{-1} and the standard deviation of the common logarithm is 0.41.

The calculated D_{life} is shown in **Figure 32** as a function of C_{soil} for three K_d values; 5, 50, and 95 percentile values. It is found that D_{life} and C_{soil} are linearly related. The safety level, however, cannot be determined based on only this relationship because the D_{life} has a variability due to the variation of the Cs-K_ds.

In accordance with the principle presented above (*The dose that each resident will get in his lifetime should be less than the dose limit*), the soil cannot be considered safe when a resident is expected to get more than the maximum dose

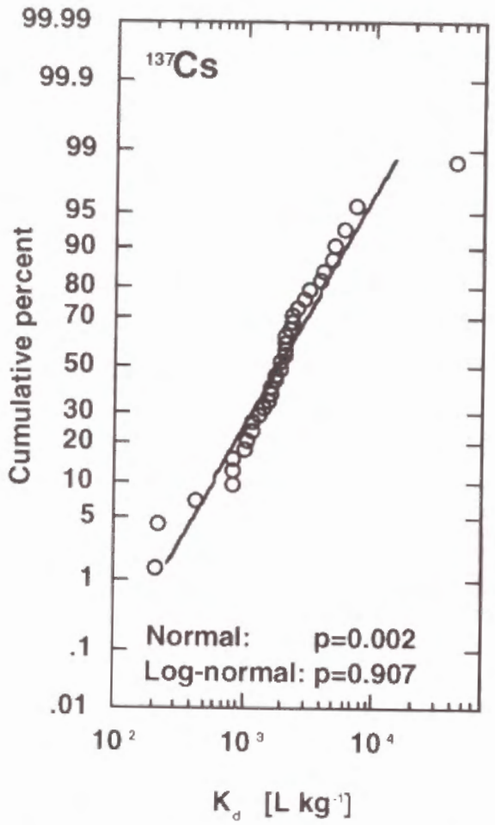

Figure 31. Probability distribution of cesium K_ds obtained for 36 agricultural soils collected in Japan.

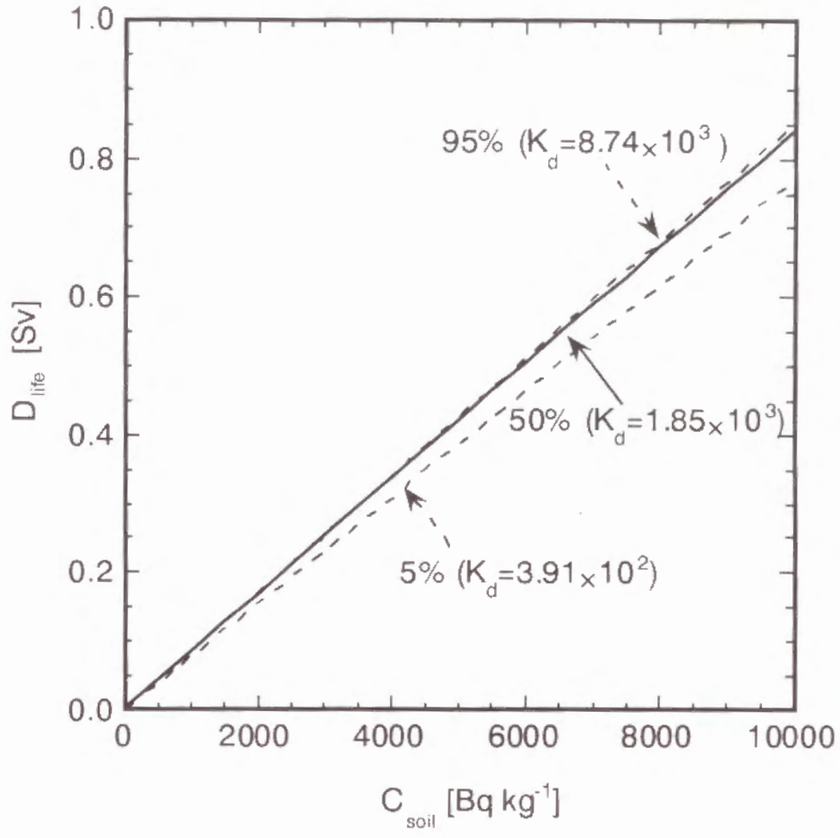

Figure 32. Predicted life-dose for a resident as a function of C_{soil}; the resident is assumed to be exposed

limit (D_{limit} [Sv]). More specifically, the maximum dose to the residents should be less than D_{limit}. However, it is difficult to estimate precisely the maximum individual dose because the environmental conditions vary to a large extent not only periodically, but also randomly. Then, the predicted number (N_{ex} [man]) of residents who will get more than the D_{limit} is calculated here by using a probabilistic technique as follows:

$$N_{ex} = P_{ex} \times N_{residents} \qquad (63)$$

where P_{ex} [-] is the probability of getting more than the D_{limit}; and $N_{resident}$, the total number of the residents [man]. The P_{ex} varies depending on the values of model parameters. Equation 63 shows that the number of residents is very important as well as the P_{ex} for each resident.

The relationship between C_{soil} and N_{ex} is shown in **Figure 33**. Here $N_{resident}$ has been assumed to be 100 [man]. Based on the principle presented, the safety level is defined as the C_{soil} when the N_{ex} is predicted to be 1. From the figure, we can determine that the safety level in this situation is 4.1×10^{3} Bq kg^{-1}. This value, however, changes depending on the variabilities of the used parameters. When the C_s-K_d has a larger variability, the slope of the curve will be more gentle and a higher safety level will be obtained. Conversely, stable parameter values will lead to a lower safety level.

5.2.2. Decision on countermeasures: cleanup or compensation

When the soil is judged to be unsafe for residence, we need to decide which countermeasure should be performed. Cleanup is not necessarily justified for responsible parties in terms of the cost-benefit consideration, although it may be requested by residents. In this decision, the principle will be

Cleanup is more advantageous than other countermeasures.

Here the advantage of each countermeasure is evaluated on the basis of its cost. As an example, cost of compensation (evacuation) is calculated and compared with that of cleanup.

The compensation period (Y_{comp} [y]) can be estimated as the time period necessary for radionuclide concentration in soil (C_{soil}) to reach the safety level for residence (C_{safe}). Then, from eq. 58, the Y_{comp} can be calculated by

$$Y_{comp} = -\frac{1}{\lambda}\ln\left(\frac{C_{safe}}{C_{soil}(0)}\right) \qquad (64)$$

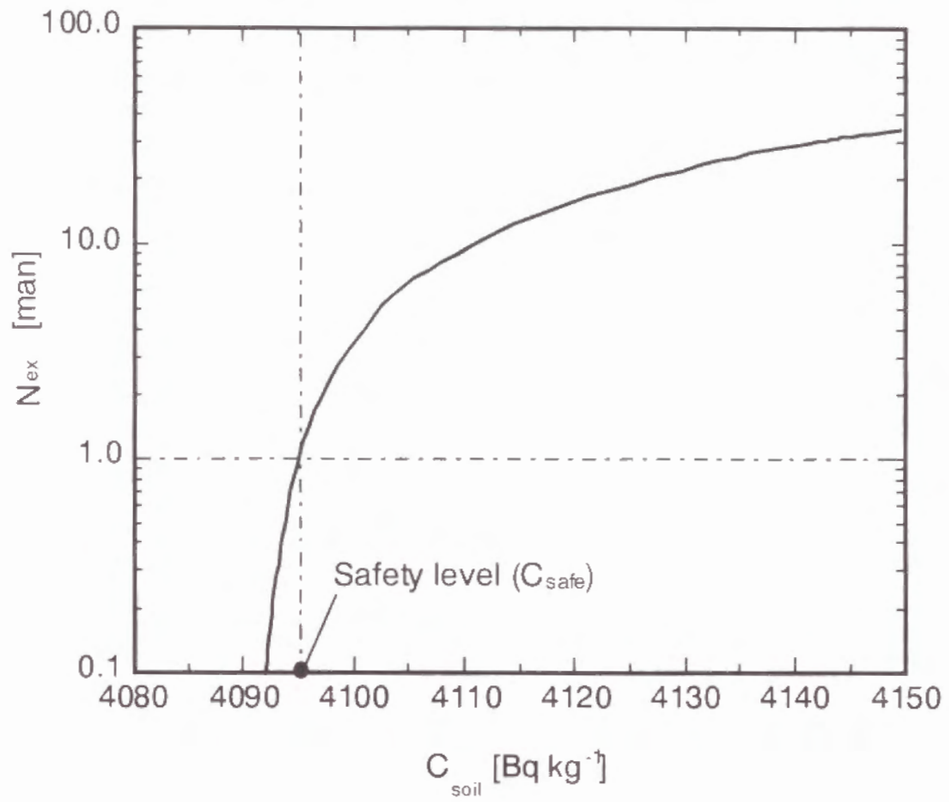

Figure 33. Predicted number of residents who will get more than 350 mSv in their lives as a function of C_{soil}.

The total compensation cost (M_{comp} [$]) is determined by

$$M_{comp} = m_{comp} \times Y_{comp} \times N_{residents} \qquad (65)$$

where m_{comp} is the annual cost for compensation per resident [$ man^{-1} y^{-1}].

The parameter values are shown in **Table 23**. The λ, effective decay constant, is calculated by eq. 60. The calculated M_{comp}s are shown in **Figure 34** as a function of C_{soil}. Cleanup is justified only when its expected cost is lower than the expected value of the M_{comp}. For instance, if the cleanup cost is less than 300 million dollars, it will be justified when the C_{soil} is greater than 20 kBq per kg. It should be noted that the expected value does not equal the median (50 percentile) value because normal type distribution is not necessarily applicable to environmental parameters such as the Cs-K_d shown in Figure 31.

5.2.3. Decision on safety of cleanup activities

Workers involved in cleanup activities are expected to be exposed to radionuclides present in the surface soil. Their health is as important as that of residents and soil cleanup should not be perfomed when the workers are predicted to experience adverse health effects. The principle for workers' safety level should be given as follows:

The dose that each worker will get in his cleanup activity should be less than the dose limit.

The extent of their risk depends on not only the contamination level, but also the time period they will be exposed. The dose to each worker (D_{worker}) can be estimated as

$$D_{worker} = \frac{A}{a_{clean} \cdot N_{worker}} \times F'_{conv} \times C_{soil}(0) \qquad (66)$$

where A is the contaminated area under consideration [m^2]; a_{clean}, the cleanup rate per worker [m^2 man^{-1} hr^{-1}]; N_{worker}, the number of workers [man]; and F'_{conv}, the conversion coefficient from radionuclide concentration in the surface soil to the dose equivalent per worker [Sv yr^{-1} per Bq kg^{-1}].

The parameter values are shown in **Table 23**. The N_{worker} is assumed to be 10 [man] and the F'_{conv} to be the same as the conversion factor for residents (F_{conv}). The D_{worker}s were calculated for three a_{clean} values (10, 50, and 100 [m^2 hr^{-1} man^{-1}]).

The relationship between the calculated D_{worker}s and C_{soil} is shown in **Figure**

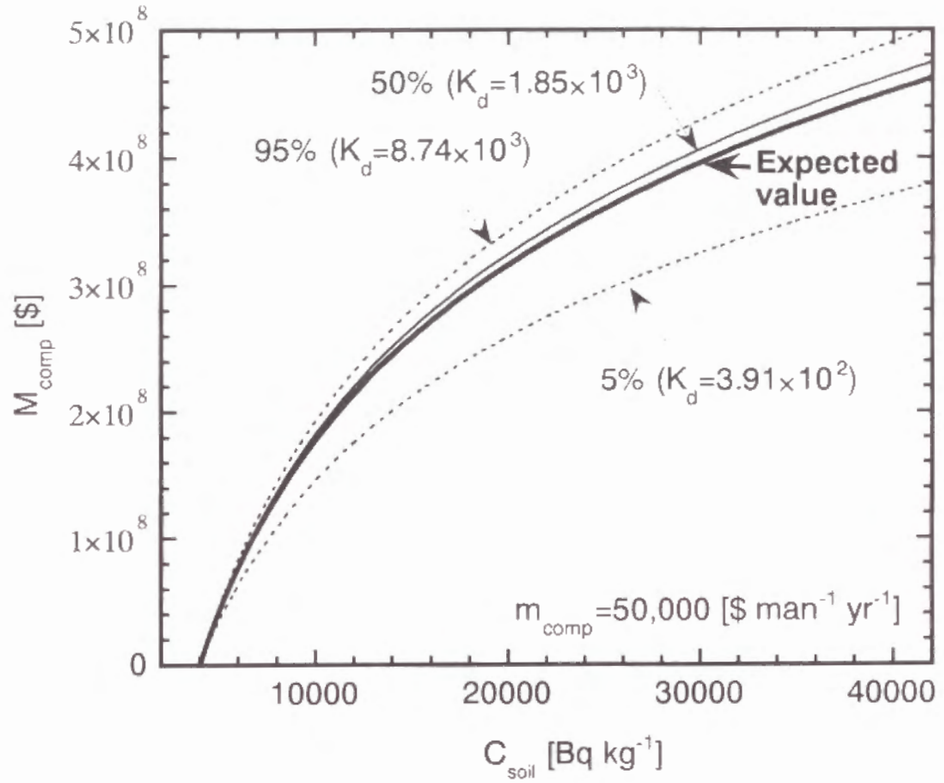

Figure 34. Estimated total compensation cost as a function of C_{soil}.

35. The 20 mSv line shown in the figure is the yearly-averaged dose limit for workers (ICRP, 1991a). If the calculated D_{worker} is expected to exceed the dose limit, access to the respective area should be prohibited. It is found that the cleanup rate (a_{clean}) is a highly sensitive factor for D_{worker}; also the number of workers (N_{worker}) has similar large sensitivity (see eq. 66). This result indicates that a larger area can be cleaned up by increasing the cleanup efficiency.

The countermeasures selected are schematically illustrated in **Figure 36** as functions of contamination level and number of residents. This figure shows that cleanup is chosen when both contamination level and number of residents are relatively high. When the number of residents is lower, the DILs for compensation and soil cleanup become higher; this DIL should be lower if the parameters have larger variability. The DIL for access-prohibition (relocation) is not affected by the number of residents, but by that of the workers. Cleanup efficiency is important to reduce the access-prohibited area.

5.3 DETERMINATION OF CULTIVATION INTERVENTION LEVEL

In case of soil contamination in agricultural lands, restriction of cultivation needs to be considered as a primary countermeasure. However, a clear principle to derive the intervention level for cultivation has not been clearly stated in previous publications (e.g. IAEA, 1986b; ICRP, 1993). Although the derived intervention level for cultivation (DILC) should be low enough to protect consumers from excessive internal irradiation, it should not be unnecessarily low because restricting cultivation will bring about serious socio-economic damages to farmers as shown after the Chernobyl accident (IAC, 1991). The DILCs should be carefully determined with the consideration that we should do more good than harm (ICRP, 1991a).

Usually, soil contamination has been evaluated from radionuclide concentrations in soil. This is based on the assumption that radionuclides are distributed in the soil and the crop at harvest with a constant ratio, T_r. The T_r values, however, have shown large variabilities as mentioned in section 2.4.2. Although we need to consider the variabilities in the determination of DILCs, such a probabilistic approach is still not yet available. This section presents a new, probabilistic method to determine the DILC.

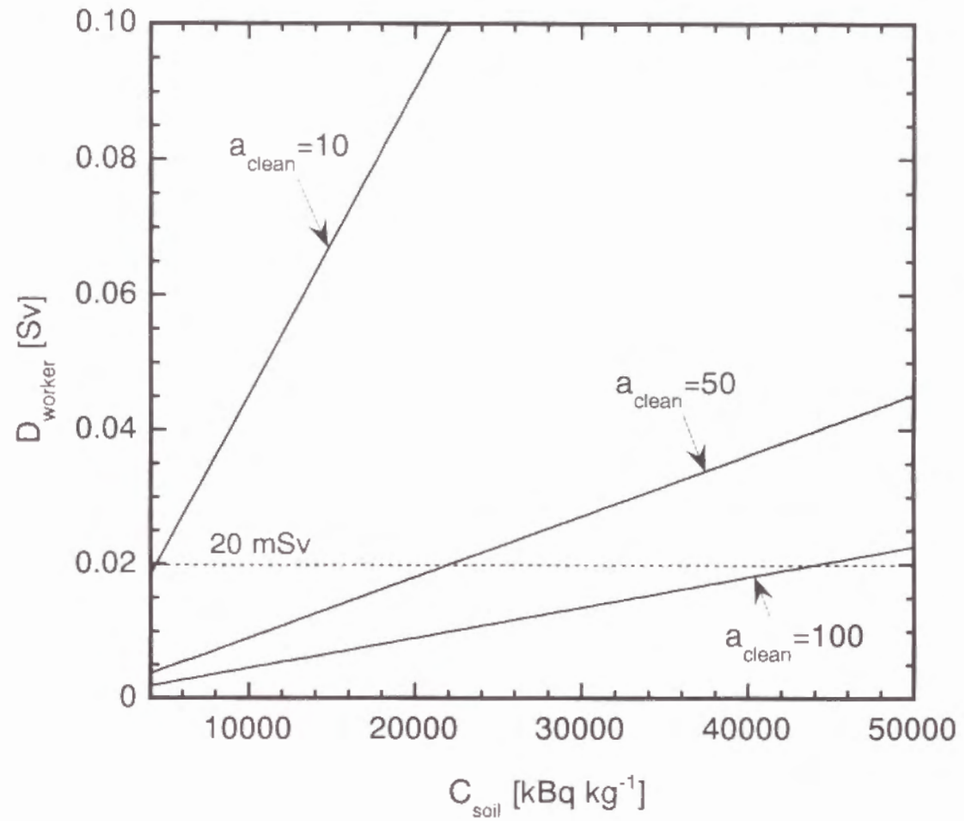

Figure 35. Predicted dose for each worker as a function of C_{soil}; the a_{clean} values in the figure are cleanup rates per worker [m^2 hr^{-1} man^{-1}].

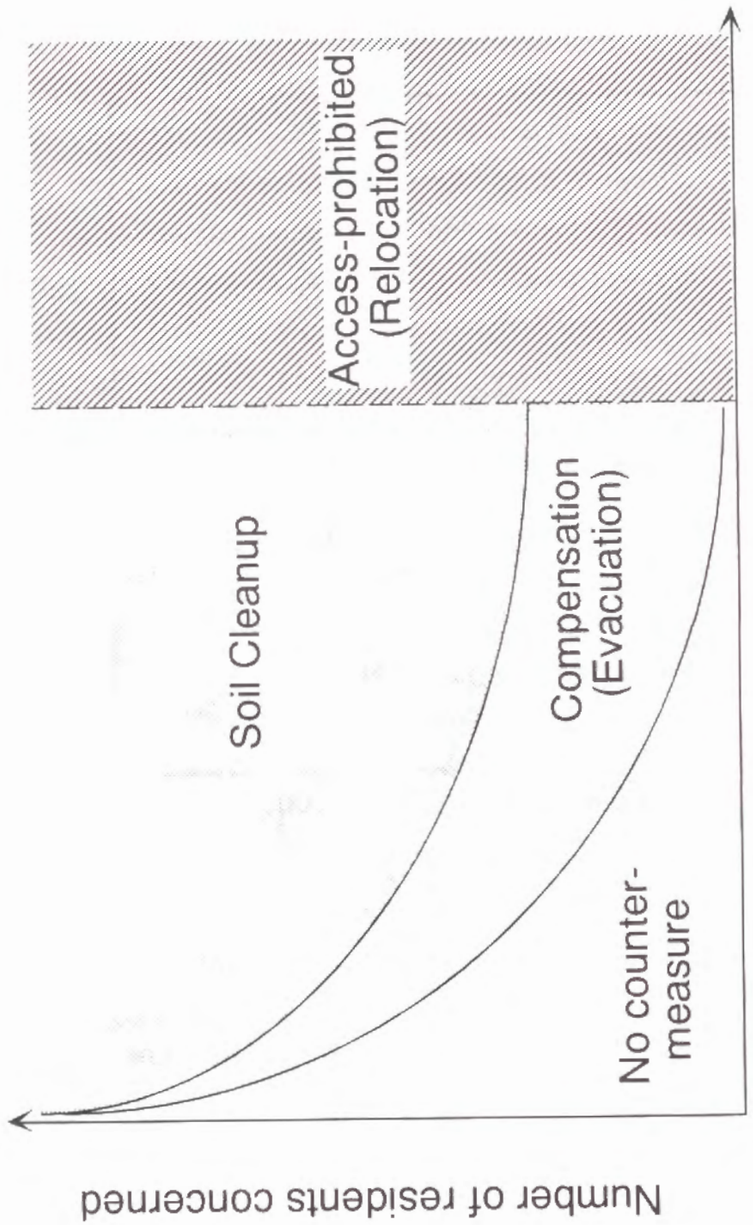

Figure 36. Schematic diagram of countermeasures related to soil contamination level and number of residents.

5.3.1 Procedure

In many radiological assessment models (e.g. Soldat and Harr, 1971; USNRC, 1977; IAEA, 1982; Muller and Prohl, 1993; Abbott and Rood, 1994), radionuclide concentration in crop edible parts at harvest (C_{crop}) [Bq kg^{-1}] is predicted by multiplying the soil-to-plant transfer factor by the concentration in soil as follows:

$$C_{crop} = T_f \cdot C_{soil} \qquad (67)$$

where C_{soil} is the radionuclide concentration in soil at harvest [Bq kg^{-1}]; and T_f, the soil-to-plant transfer factor [Bq kg-crop^{-1} per Bq kg-soil^{-1}]. The annual radionuclide intake by a consumer (I_y) [Bq y^{-1} man^{-1}] can be calculated by

$$I_y = I_c \cdot C_{crop} = I_c \cdot T_f \cdot C_{soil} \qquad (68)$$

where I_c is the annual crop consumption by a consumer [kg y^{-1} man^{-1}].

Although the DILC should be deterministic, the calculated I_y is associated unavoidably with some uncertainty due to the variation of the I_c and the T_f. For the purpose of protecting consumers' health, the DILC should be as low as possible. As a result, however, farmers will suffer from excessive socio-economic damages. To satisfy the needs of both consumers and farmers, a clear and persuasive principle needs to be established. Then, this study presents the principle as follows:

'No farmer makes a consumer exceed the limit of intake'

Although this statement seems to stress a farmer's responsibility for consumers' health, it can be also expressed as *'cultivation is allowed for the farmer who makes no consumer exceed the limit of intake'*. Then this principle is expected to balance consumers' health risk and farmers' socio-economic needs. Based on the principle, cultivation will be restricted according to the order of crop yield. As a result, the farmer having a small yield can avoid being a target of intervention; the number of farmers who experience socio-economic disruption can be minimized.

Here it is assumed that variation of the parameters involved reflects that of the radionuclide intake by consumers. Accordingly, the number of consumers (N_{ex} [man]) who will exceed the ALI_c per farmer is calculated as

$$N_{ex} = P_{ex} \cdot N_c \qquad (69)$$

where P_{ex} is the probability of exceeding the ALI_c ($I_y > ALI_c$) for a consumer; N_c, the number of consumers per farmer [man]. The P_{ex} changes depending on the variation characteristics of the parameters.

Based on the principle proposed, the N_{ex} should be less than one man. That is, cultivation is permitted when

$$N_{ex} < 1 \qquad (70)$$

The radionuclide concentration in soil when the N_{ex} is equal to 1 is defined as the DILC.

In this study, the parameter values except T_f are assumed to be constant although their variational characteristics are unclear. Hence the N_c in eq.69 can be calculated by

$$N_c = \frac{Y_c}{I_c} \qquad (71)$$

where Y_c is the annual crop yield per farmer [kg y^{-1}]. From eqs.69-71, it is suggested that a farmer producing larger amounts of a crop (Y_c) is intervened with earlier. As a result, the principle proposed above minimizes the number of farmers who must be restricted for cultivation.

This section presents sample procedures to derive cultivation intervention levels of ^{90}Sr and ^{137}Cs for two crops (cabbage and spinach) and two soil types (loam and sand). The annual limit of intake for workers (ALIs) (ICRP,1979) has already been given for major radionuclides in accordance with ICRP recommendations (ICRP, 1991b). Here the annual limit of intake for consumers (ALI_c) is defined as one-tenth the quantity of the ALI because the averaged dose limit for workers is ten times higher than that for the general public (ICRP, 1991a). The ALI [Bq y^{-1} man^{-1}] for ^{90}Sr is 6.0×10^5 and that for ^{137}Cs is 1.0×10^6 when the fractions of Sr and Cs reaching the body fluids following ingestion are assumed to be 0.3 and 1, respectively (ICRP, 1991b). The variation of T_fs is estimated using the data collected by the International Union of Radioecologists (IUR, 1987). The probability distributions of T_fs for ^{90}Sr and ^{137}Cs are shown in **Figure 37**. These are approximated not by a normal distribution type, but by a log-normal one (Yasuda and Uchida, 1994). Their geometric means and standard deviations of the common logarithm (SDCLs) are shown in **Table 24**. The magnitude of the geometric means is in the order of sand, loam+sand, and loam for all cases. For I_c (MHW, 1993) and Y_c (MAFF, 1992a), the averaged values in Japan are adapted as shown in **Table 25**; in the calculation, these are converted to the values on a dry-weight basis assuming that the water content ratio is 0.9 (Ng et al., 1982).

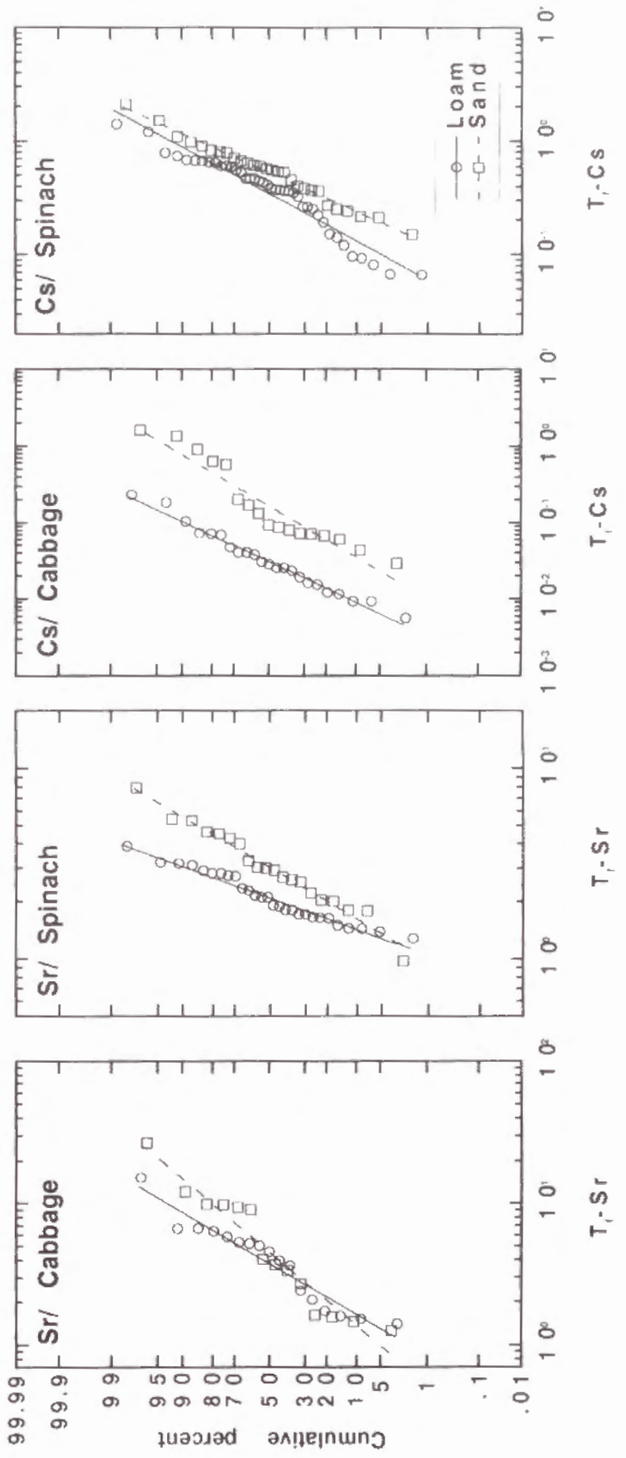

Figure 37. Probability distributions of soil-to-plant transfer factors summarized by International Union of Radioecologists (IUR 1987).

Table 24. Statistical values of soil-to-plant transfer factors in the IUR report (1987) on a dry weight basis.

crop	soil	Sr			Cs		
		n	geometric mean	std. dev. com. log. [a]	n	geometric mean	std. dev. com. log. [a]
cabbage	loam+sand	31	4.10	0.347	40	0.063	0.594
	loam	17	3.77	0.289	23	0.031	0.218
	sand	14	4.55	0.414	17	0.166	0.294
spinach	loam+sand	48	2.43	0.184	68	0.409	0.320
	loam	28	2.08	0.130	40	0.342	0.333
	sand	20	3.00	0.210	28	0.527	0.269

[a] The standard deviation of the common logarithm.

Table 25. Parameters relating to agricultural production and consumption.

crop	cultivated area [a] [m^2]	annual crop intake per consumer [b] I_c [kg y^{-1} man^{-1}]	annual crop yield per farmer [a] Y_c [kg y^{-1}]	number of consumers per farmer N_c [man]
cabbage	4.04×10^{8}	7.48	1.16×10^{5}	1.55×10^{4}
spinach	2.73×10^{8}	5.22	4.26×10^{4}	8.16×10^{3}

[a] On fresh weight basis (MAFF 1990).
[b] On fresh weight basis (MHW 1993).
[c] Averaged value in Japan, which was calculated by dividing total crop yield by total number of farmers.

5.3.2 Results and Discussion

The relationships between the C_{soil}s and the I_ys are shown in **Figure 38** for ^{90}Sr and in **Figure 39** for ^{137}Cs. These are shown for three T_f values: 5, 50, and 95 percentiles. It is found that the variation of I_y is very large. Derived N_{ex}s for ^{90}Sr and ^{137}Cs as a function of C_{soil} are plotted in **Figure 40** for ^{90}Sr and **Figure 41** for ^{137}Cs. Although the N_{ex}s increase with C_{soil} in all cases, the relationships are not linear; they gradually approach the maximum value, i.e. total number of consumers. The slopes tend to be more gentle in sand than in loam, which is due to the difference in their T_f distributions (Table 24).

The DILC values are shown in **Table 26**. Larger DILC is derived when the T_f has a smaller variability represented by the SDCL. The order of magnitude of the DILCs does not necessarily correspond to that of the geometric means of T_fs. In

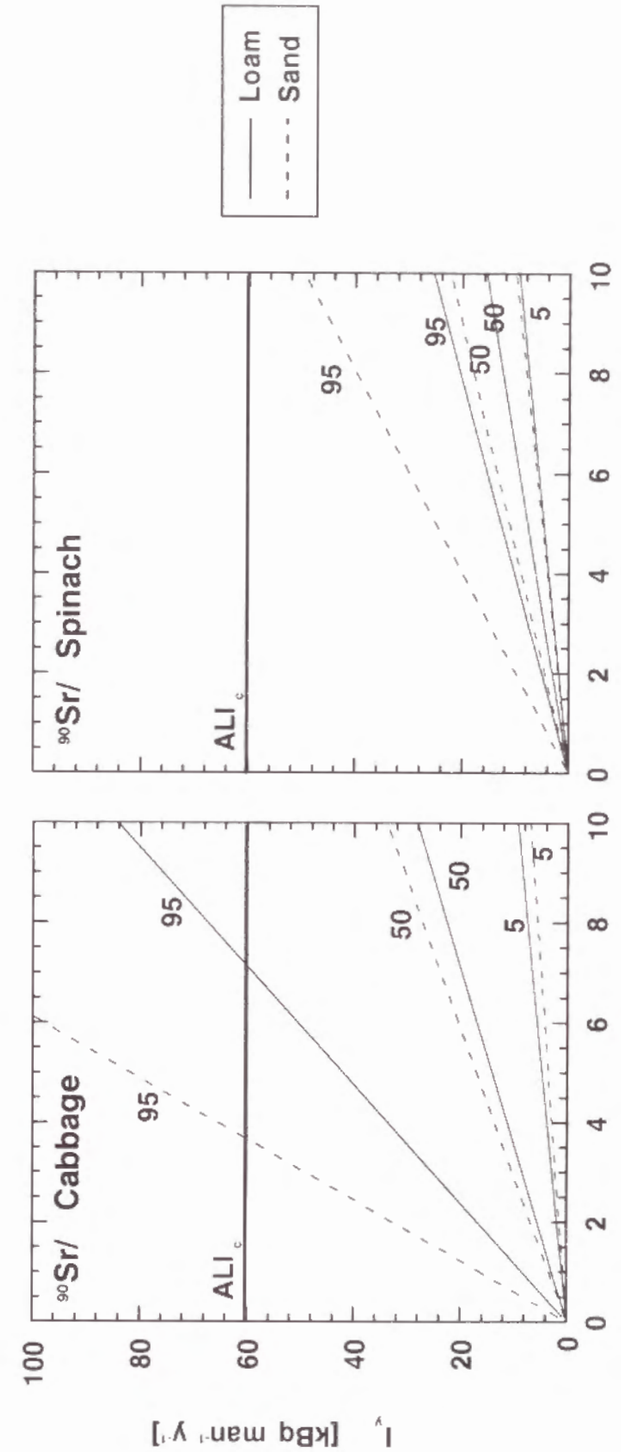

Figure 38. Plots of calculated annual intake of ^{90}Sr per consumer (I_y) versus the concentration in soil (C_{soil}).

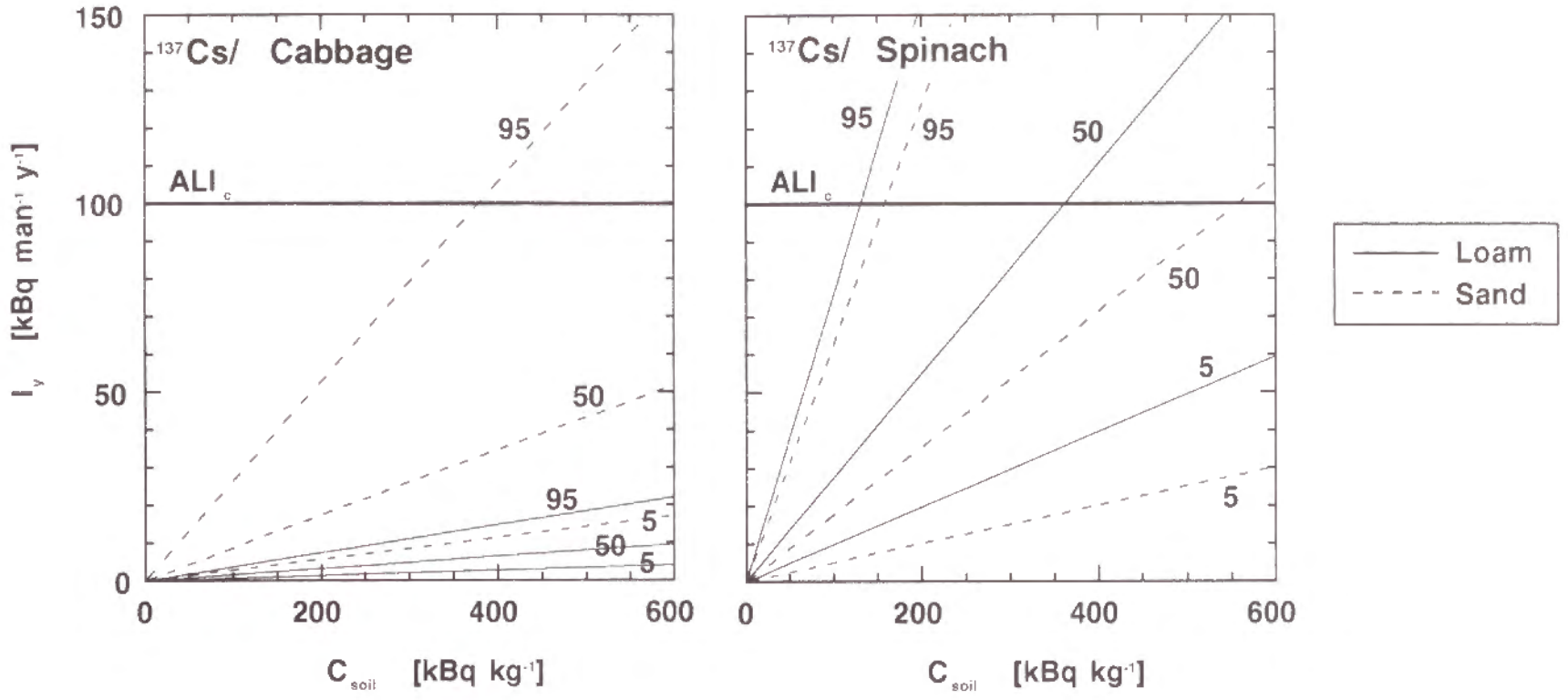

Figure 39. Plots of calculated annual intake of ^{137}Cs per consumer (I_y) versus the concentration in soil (C_{soil}).

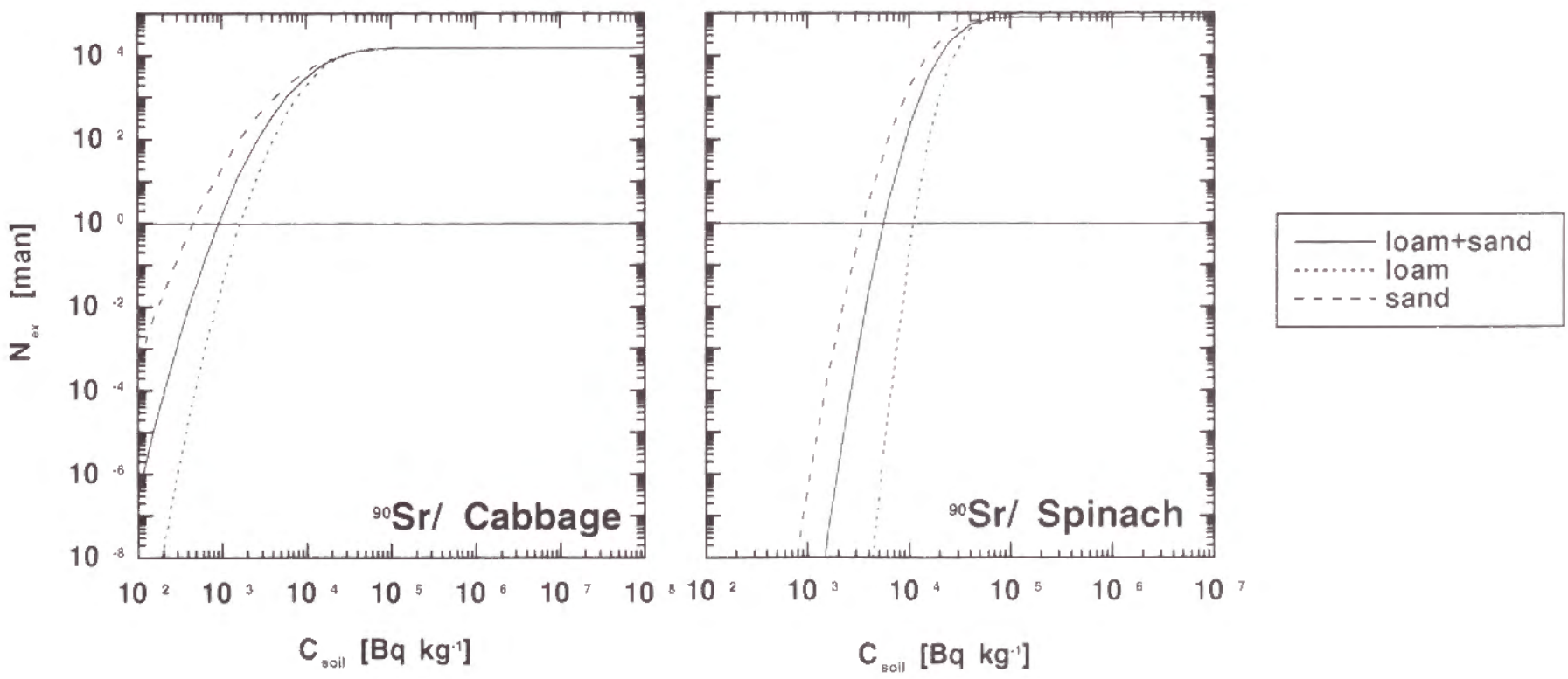

Figure 40. Plots of estimated number of consumers who would ingest ^{90}Sr above the ALI versus the ^{90}Sr concentration in soil.

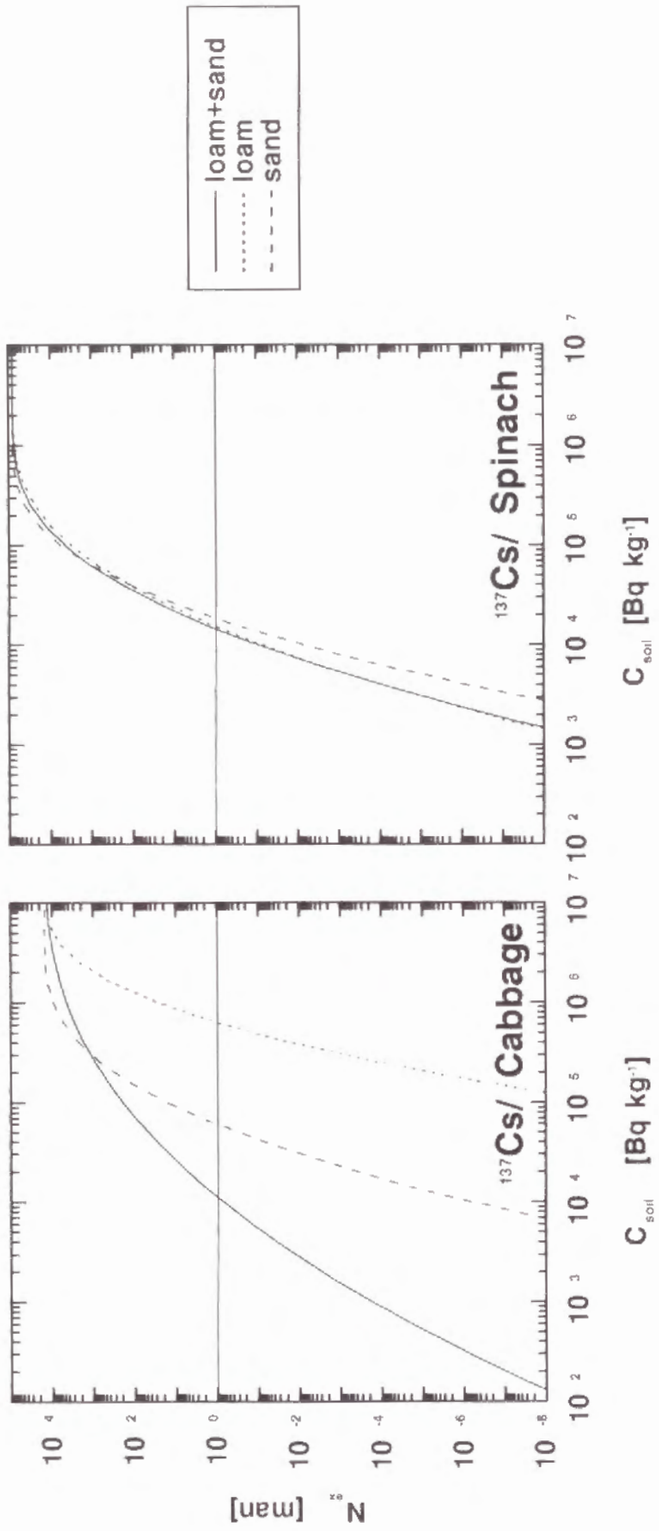

Figure 41. Plots of estimated numbers of consumers who would ingest ^{137}Cs above the ALI versus the ^{137}Cs concentration in soil.

Table 26. Derived intervention levels for cultivation (DILCs) calculated from the values in Tables 24 and 25.

crop	soil type	DILC [Bq kg^{-1}]	
		^{90}Sr	^{137}Cs
cabbage	loam+sand	9.19×10^{2}	1.13×10^{4}
	loam	1.67×10^{3}	6.31×10^{5}
	sand	4.59×10^{2}	6.03×10^{4}
spinach	loam+sand	7.92×10^{3}	2.09×10^{4}
	loam	1.56×10^{4}	2.21×10^{4}
	sand	4.98×10^{3}	2.66×10^{4}

[a] The standard deviation of the common logarithm.

case of ^{137}Cs contamination, the DILCs for cabbage and spinach are larger in sand than in loam+sand; the oppsite results would be obtained if only the geometric means were considered. These results suggest that quantification of parameter variability is primarily important when deriving suitable DILCs based on the principle presented in this study. If a parameter has a large uncertainty, the DILC should be small enough to protect consumers from the potential health risk. As a result, cultivation would be excessively restricted. Conversely, if the parameter is reliably determined, a higher DILC could be set and lives of fewer farmers would disrupted. More effort to clarify the variability of each parameter is needed rather than only showing the representative value.

After determining the DILCs, it is necessary to discuss their necessity in comparison with the levels of other countermeasures such as evacuation and relocation. As an example, the DILCs are compared with the relocation levels (DILRs) in the following. The DILRs of ^{90}Sr and ^{137}Cs deposited on the ground are reported to be 3×10^{8} [Bq m^{-2}] and 4×10^{6} [Bq m^{-2}], respectively (IAEA, 1986). Assuming both radionuclides are uniformly distributed in the surface soil having a density of 240 [kg m^{-2}] (USNRC 1977), the DILCs obtained here (Table 26) are revised as 6.8×10^{5} - 3.8×10^{7} [Bq m^{-2}] for ^{90}Sr and 1.3×10^{7} - 1.2×10^{9} [Bq m^{-2}] for ^{137}Cs. Comparing these values, the DILC for ^{90}Sr, which is much smaller than the relocation level, is necessary for protecting consumers. The DILC for ^{137}Cs, however, is less important because farmers would be relocated at lower levels.

It should be noted that the DILCs obtained here are specimen values. They should be derived for each situation using the values locally obtained. The DILCs

may possibly differ when using local values for the parameters. The DILCs would be smaller when the soil is contaminated by other radionuclides. Dilution through market flow and food preparation process would reduce the radionuclide intake by each consumer, which will lead to higher DILCs being set. Additionally, DILCs might not be necessary when another countermeasure such as relocation is justified. The priority of possible countermeasures must be carefully determined, based on information collected site-specifically with full consideration of social and economic factors. The method presented here is expected to be effectively applied to such a problematic decision-making process.

5.4 APPLICATION OF THE FLEXIBLE TF MODEL TO ENVIRONMENTAL IMPACT ASSESSMENTS

The model (eq.53) which predicts the T_f value using selected parameters corresponding to the growth conditions was developed (see section 4.4). It has a good possibility for estimating the suitable T_f in actual fields where the environmental conditions vary greatly. In this section, the model is applied to a numerical simulation of ^{90}Sr transfer from soil to crop and its effectiveness is discussed through the sensitivity analyses under simulated conditions in comparison with the method using a constant T_f.

5.4.1. Models and Conditions

As mentioned in Chapter 2, chronological change of ^{90}Sr concentration in surface soil can be described as an exponential type equation (IAEA, 1982; Peterson, 1983; NCRP, 1984). That is, when the ^{90}Sr has been momentarily deposited onto the soil surface in an agricultural field, the ^{90}Sr concentration in the soil (C_{soil}) [kBq kg^{-1}] is given by

$$C_{soil}(t) = C_{soil}(0)\cdot\exp(-\lambda\ t) \qquad (72)$$

where $C_{soil}(0)$ is the initial ^{90}Sr concentration in the surface soil [kBq kg^{-1}]; t, the time [y]; and λ, the total decay constant [y^{-1}]. The λ can be described by three main three factors as follows (NCRP, 1984):

$$\lambda = \lambda_d + \lambda_i + \lambda_c \qquad (73)$$

where λ_d is the radioactive decay constant [y^{-1}]; λ_i, the downward infiltration rate [y^{-1}]; and λ_c, the removal rate by crops [y^{-1}].

Here the λ_d is given by the following equation considering water uptake by plants:

$$\lambda_i = \frac{V_w}{W_{soil}\cdot(\theta_l + K_d)} \qquad (74)$$

where V_w is the water infiltration rate per unit area [L m^{-2} y^{-1}]; W_{soil}, the amount of surface soil per unit area [kg m^{-2}]; θ_l, the soil water content by weight [L kg^{-1}]; and K_d, the soil-solution distribution coefficient [L kg^{-1}].

Accordingly, when $\theta_l << K_d$, the λ is described by

$$\lambda = \frac{\ln 2}{T_{Sr}} + \frac{V_w}{W_{soil}\cdot K_d} + \frac{T_f\cdot Y_c}{W_{soil}} \qquad (75)$$

where T_{Sr} is the half life of ^{90}Sr (=28.8y); and Y_c, the annual crop yield per unit area [kg m^{-2}]. Here all of the ^{90}Sr absorbed by roots is assumed to translocate into the edible parts; this assumption is reasonable for leafy vegetables (NCRP, 1984).

Therefore the concentration in the crop edible parts (C_{crop}) [kBq kg^{-1}] is predicted by multiplying the C_{soil} by the T_f:

$$C_{crop}(t) = T_f\cdot C_{soil}(0)\cdot\exp\left\{-\left(\frac{\ln 2}{T_{Sr}} + \frac{V_w\cdot EC}{W_{soil}\cdot 2.1\cdot CEC} + \frac{T_f\cdot Y_c}{W_{soil}}\right)\cdot t\right\} \qquad (76)$$

According to the results of the section 4.4 (eq.57), the T_f of strontium for *Brassica campestris* can be given by

$$T_f = \frac{a\cdot(EC)^b\cdot T_c}{CEC} \qquad (77)$$

where a and b are constants; EC is the electrical conductivity in the solution [mS cm^{-1}], CEC is the cation exchange capacity of the soil [mmol(+) 100g^{-1}]. The a and b values obtained for *Brassica campestris* were 0.63 and 0.28, respectively. On the basis of the mass balance of water in surface soil, the V_w is

$$V_w = V_b - T_c\cdot Y_c \qquad (78)$$

where V_b is the water infiltration velocity per unit area without plants [L m^{-2} y^{-1}]; and T_c, the transpiration coefficient [L kg^{-1}]. Substituting eqs.77 and 78 into eq.76, the C_{crop} can be given by

$$C_{crop}(t) = \frac{a\cdot(EC)^b\cdot T_c}{CEC}\cdot C_{soil}(0)\cdot\exp\left\{-\left(\frac{\ln 2}{T_{Sr}} + \frac{(V_b - T_c\cdot Y_c)\cdot EC}{2.1\cdot CEC\cdot W_{soil}} + \frac{a\cdot(EC)^b\cdot T_c\cdot W_y}{CEC\cdot W_{soil}}\right)\cdot t\right\} \qquad (79)$$

Arranging eq.79 gives

$$C_{crop}(t) = \frac{a \cdot (EC)^b \cdot T_c}{CEC} \cdot C_{soil}(0) \cdot \exp\left\{-\left(\frac{\ln 2}{T_{Sr}} + \left\{\frac{V_b \cdot EC}{2.1} + \left(a \cdot (EC)^b - \frac{EC}{2.1}\right) \cdot T_c \cdot Y_c\right\} \cdot \frac{1}{CEC \cdot W_{soil}}\right) \cdot t\right\} \quad (80)$$

In this simulation, the initial ^{90}Sr concentration in the surface soil is assumed to be a normal distribution of two dimensions as follows:

$$C_{soil}(0) = \frac{1}{W_{soil}} \times \frac{M}{2\pi\sigma^2} \cdot \exp\left(-\frac{X^2 + Y^2}{2\sigma^2}\right) \quad (81)$$

where M is the total amount of ^{90}Sr released from the source [kBq]; σ, the standard deviation [m]; X, the distance from the source on a horizontal scale [m]; and Y, the distance from the source on a vertical scale [m].

The parameters used for the simulation are shown in **Table 27**. The V_b is the mean of water infiltration velocities in references (Hoffman and Baes, 1979); Y_c, the vegetable yield areally averaged in Japan (MAFF, 1992a); W_{soil}, the areal density of plowed layer soil (USNRC, 1977); T_c, the mean of the observations for *Brassica campestris* (section 4.4). The M is given as the 10 % of the amount of ^{90}Sr released in the Chernobyl accident (UNSCEAR, 1988). The distribution of the initial ^{90}Sr concentration is shown in **Figure 42**. The north region from the source is assumed to be ocean and ^{90}Sr deposited onto this area is not considered. Here about 45 % of the M is calculated to deposit onto the illustrated area of 200 km^2 (X: 20 km × Y: 10 km).

5.4.2. Index of risk evaluation

The safety limit of radionuclide concentration in the crop (C_{limit}) [Bq kg^{-1}] can be led from the annual limit of intake (ALI) [kBq man^{-1} y^{-1}] presented by ICRP

Table 27. Parameter values used for the simulation of ^{90}Sr transfer.

V_b [L/m^2/y][a]	Y_c [kg/m^2/y][b]	W_{soil} [kg/m^2][c]	T_c [L/kg][d]	EC [mS/cm]	M [kBq]	σ [km]
740	3.4	240	36.0	1.0	2.0×10^{13}	6.0

[a] Taken from Hoffman and Baes (1979).
[b] The 1990 annual yield of leafy vegetables in Japan (MAFF, 1992a).
[c] Taken from USNRC (1977).
[d] The averaged value for *Brassica campestris* (see section 4.4).

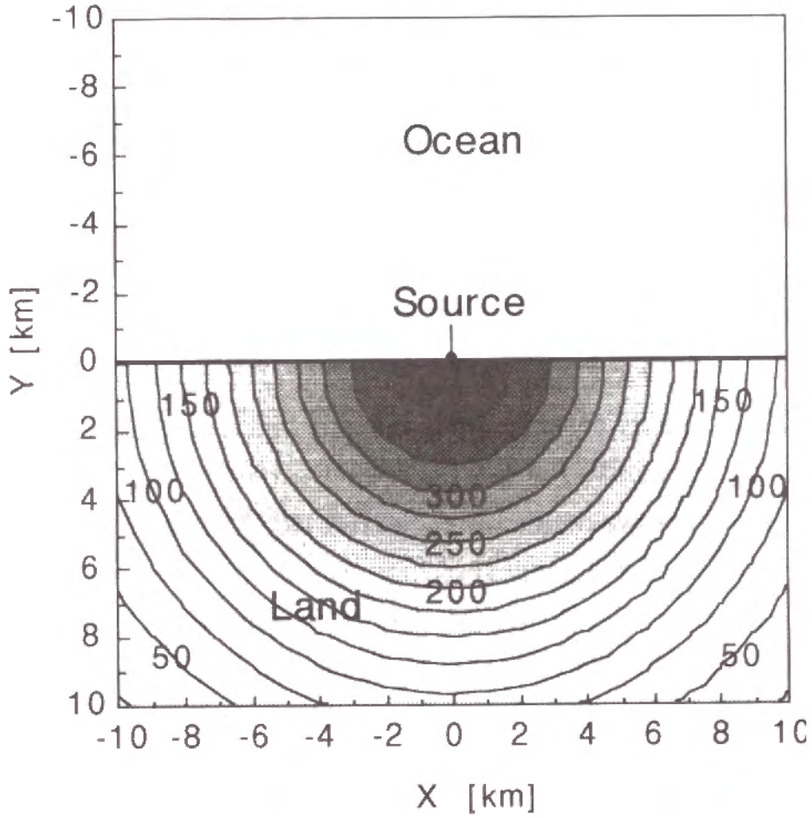

Figure 42. Initial distribution of ^{90}Sr concentration in surface soil; the values in the figure shows the concentration levels [kBq kg^{-1}].

(1991b) as follows:

$$C_{limit} = \frac{ALI}{I_d \times 365} \qquad (82)$$

where I_d is the daily intake of the crop [kg man^{-1} d^{-1}].

Cultivation should be restricted for the agricultural field where the produced crops have larger concentration than the C_{limit}. Therefore, the standard of the restriction is defined as follows:

$$C_{soil} \geq \frac{C_{limit}}{T_f} \qquad (83)$$

According to eq.72, the restriction period (Y_R) [y] is calculated by

$$Y_R = -\frac{1}{\lambda_{di}} \times \ln\left(\frac{1}{C_{soil}(0)} \cdot \frac{C_{limit}}{T_f}\right) \qquad (84)$$

where λ_{di} ($= \lambda_d + \lambda_i$) is the decay constant when the cultivation is restricted [y^{-1}].

The farmers restricted for cultivation will experience economic hardship due to the loss of crop yields. Then we define the social risk (R_{soc}) as the crop yield lost by the restriction. The avoidable medical risk (R_{med}) is defined as the cancer risk avoided by the restriction, in other words, the risk which consumers would get if the agricultural activities were continued. These index values are expressed by the following equations:

$$R_{soc} = \int_s Y_R \cdot Y_c \cdot ds \qquad (85)$$

$$R_{med} = \int_s \left\{ \int_0^{Y_R} Y_c \cdot C_{crop}(t) \cdot dt \right\} \cdot ds \times f_{DE} \times f_{CR} \qquad (86)$$

where s means area [m^2]; f_{DE}, the ingestion dose equivalent [Sv kBq^{-1}]; f_{CR}, the cancer risk by internal radiation of 1 Sv [man Sv^{-1}]. The $C_{crop}(t)$ in eq.86 is calculated by eq.80.

The values used for the risk estimation are shown in **Table 28**. The ALI is the annual limit on intake of ^{90}Sr for workers (ICRP, 1991b); I_d, the averaged daily consumption of leafy vegetables for a Japanese (MAFF, 1992b); f_{DE}, the ingestion dose equivalent for an adult (ICRP, 1989); and f_{CR}, the fatal cancer risk for the whole population (ICRP, 1991a).

Table 28. Parameter values used for the simulated risk evaluation.

ALI [kBq/man/y][a]	I_d [kg/man/y][b]	f_{DE} [Sv/kBq][c]	f_{CR} [man/Sv][d]
6.0×10^{2}	47.5	3.5×10^{-5}	5.0×10^{-2}

[a] ICRP (1990b).
[b] MAFF (1992).
[c] ICRP (1989).
[d] ICRP (1990a).

5.4.3. Calculated results

Changes of the relative concentrations in the soils ($C_{soil}(t)/C_{soil}(0)$) are shown in **Figure 43** for three CECs: low (7 mmol(+) 100g^{-1}), middle (13 mmol(+) 100g^{-1}), and high (26 mmol(+) 100g^{-1}) CEC. These three CECs are 10, 50, and 90 percentile values, respectively, of those obtained for 36 agricultural soils (Table 9). The C_{soil} decreases more rapidly in the lower CEC soil. If the contamination level is evaluated using only C_{soil}, the soil having a higher CEC is restricted from being cultivated for a longer period.

In **Figure 44**, change of C_{crop} is plotted as a function of time for each soil CEC. Though the C_{crop} decreases most rapidly in lower CEC as the C_{soil}, its value is highest in lowest CEC initially. This suggests that the contamination level of an agricultural field cannot be judged by only the C_{soil}; its safety level should be deter-

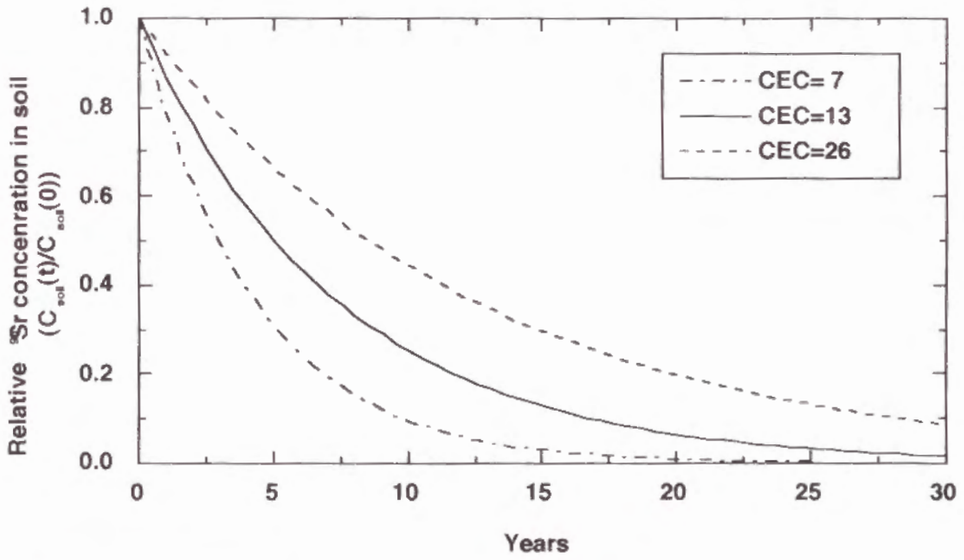

Figure 43. Predicted change of ^{90}Sr concentration in soil of three CECs [mmol(+) 100g^{-1}-soil] when cultivation is restricted.

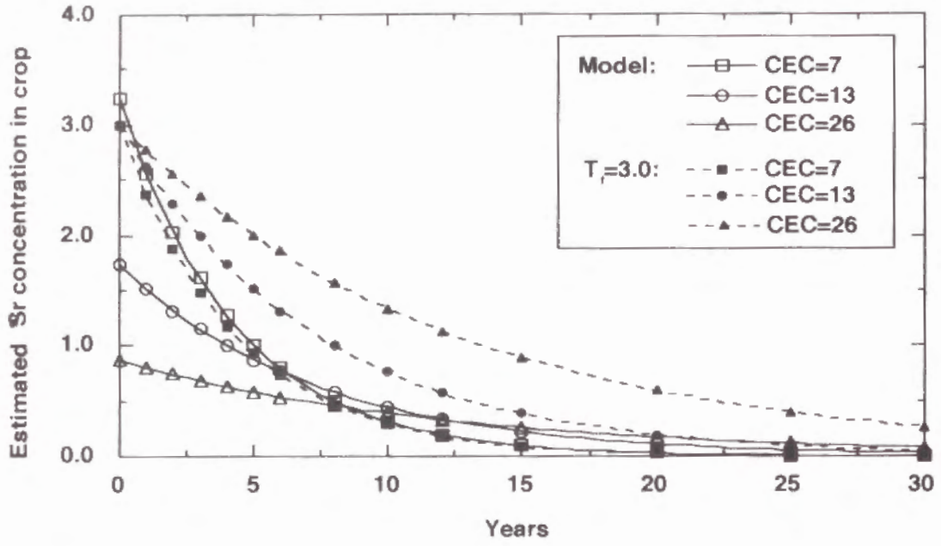

Figure 44. Predicted change of ^{90}Sr concentration in crop for three CECs [mmol(+) $100g^{-1}$-soil] in comparison between two methods: the proposed model and a constant T_f (=3.0).

mined flexibly on the basis of the characteristics of ^{90}Sr behavior in each soil-plant system.

The C_{soil} and the C_{crop} calculated show different relationships with soil CEC (**Figure 45**). Though both concentrations decrease rapidly in low CEC, the curves of both concentrations as a function of CEC show inverse relationships initially. This is because the T_f becomes larger with decreasing CEC as can be predicted by eq.77. As a result, an apparent non-linear relationship between the C_{soil} and the C_{crop} appears (**Figure 46**). This tendency agrees with that found in the obsevations shown in Figure 1 although the nuclide is different. Then it is considered that we would get larger health risk from the soil of lower CEC in a initial stage providing that the cultivation would not be restricted. In other words, rapid countermeasures such as restriction of cultivation is considered to be more effective for the soil of low CEC so that large medical risk may be avoided. Consquently, in the case of ^{90}Sr contamination, the critical area for which the restriction should be carried out with priority should be the site having a low CEC.

The estimated risks are shown in **Figure 47** as a function of distance from the source under consideration. The R_{med} is smaller in higher CEC soil though the R_{soc} is larger, which can be explained by the results shown in Figure 45. Although both the R_{soc} and R_{med} are increasing with the M, the relationship is not

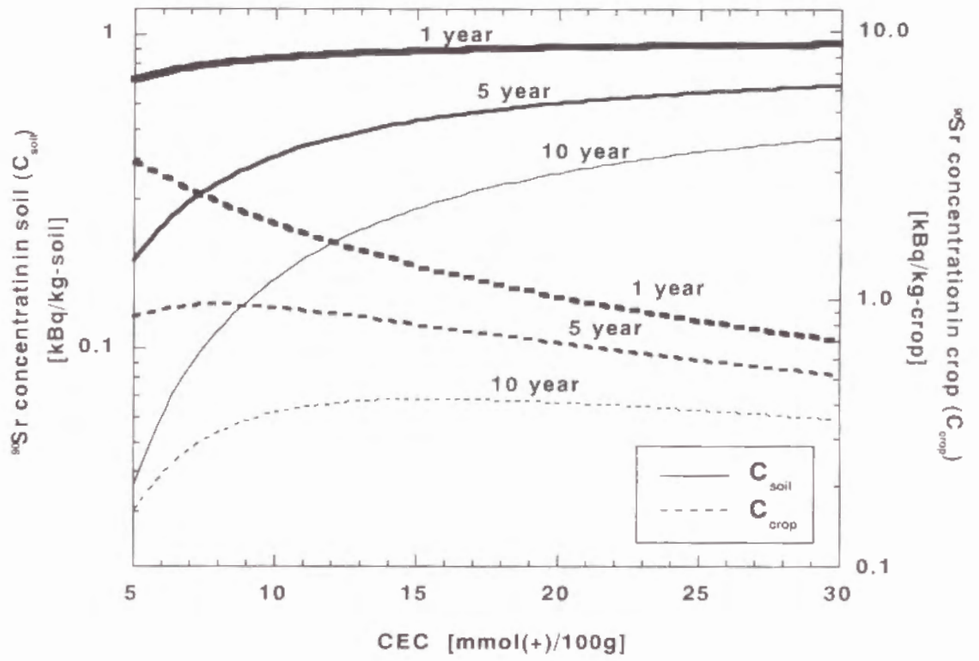

Figure 45. Plots of soil CEC versus calculated ^{90}Sr concentration in soil and that in crop when the initial C_{soil}=1 [kBq kg^{-1}].

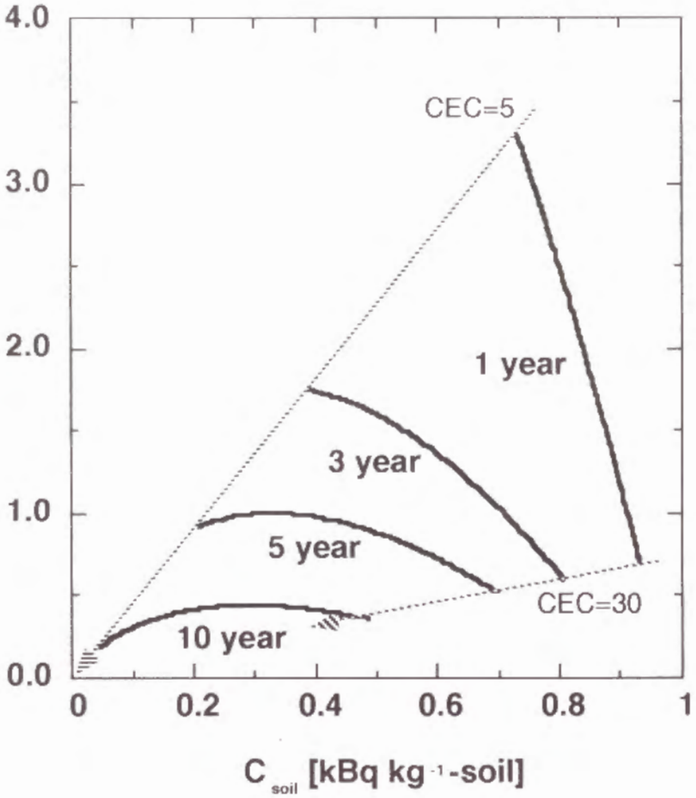

Figure 46. Relationship between C_{soil} and C_{crop} calculated for the range of CEC from 5 to 30 mmol(+) $100g^{-1}$-soil.

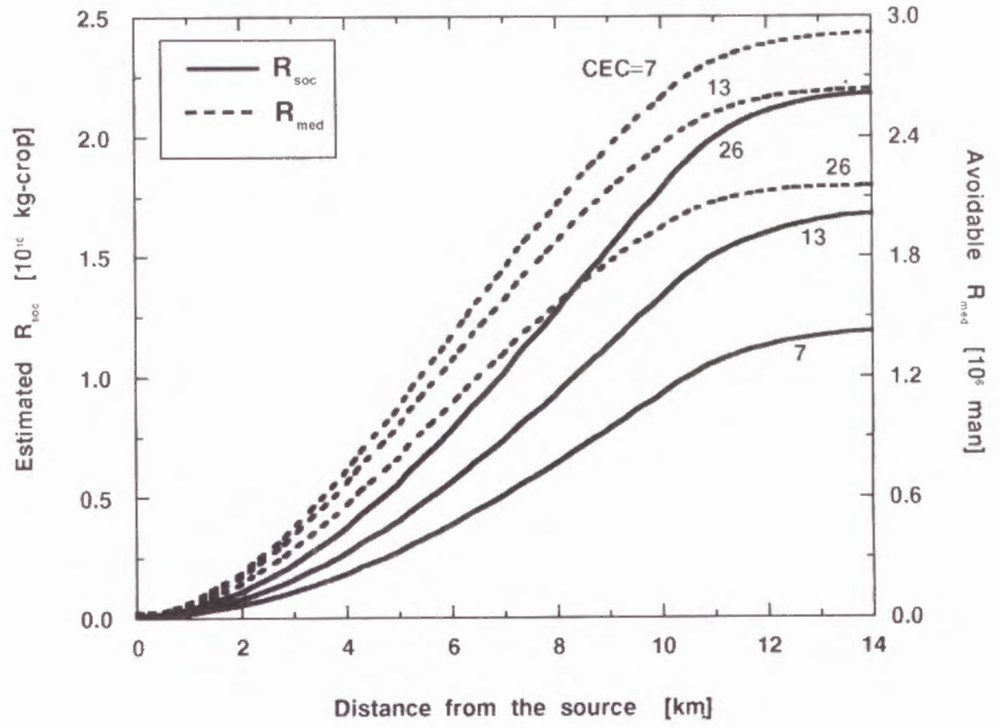

Figure 47. Estimated social risk (R_{soc}) and avoidable medical risk (R_{med}) as a function of distance from the source; the values in the figure are the CECs [mmol(+) $100g^{-1}$-soil].

linear but logistic; the plateaus are found for all cases at more than 10 km distance from the source, where the contamination seems not so severe.

5.4.4. Sensitivity analyses

In this section, sensitivities of some parameters on the R_{soc} and the R_{med} in the area of 200 km^2 (X: 20 km × Y: 10 km) are examined in numerical simulations. Here four parameters are chosen; they are released amount of ^{90}Sr (M), the water infiltration velocity under restriction (V_h), the transpiration coefficient (T_c), and the electrical conductivity in soil solution (EC). The risks are calculated for three soil CECs: 7, 13, and 26 [mmol(+) 100g^{-1}] which were the same values used in the section 5.3.

Effects of released amount (M) In **Figure 48**, relationships between Ms and calculated risks are shown for three CEC soils. Though the R_{med} is almost proportional to the M, the R_{soc} is not. This is because the area under consideration is limited within 10 km × 10 km; the R_{soc} would be proportional to the M if an infinite area were considered.

Effects of water infiltration velocity (V_b) It is expected that the contamination in surface soil would decrease rapidly with heavy rainfall, that is, large V_b. This idea is confirmed in the calculation results shown in **Figure 49**. Both R_{soc} and R_{med} decrease exponentially with the V_b. Accordingly water infiltration (rainfall-evaporation) is considered to be one of the most sensitive factors on these risks.

Effects of transpiration coefficient (T_c) T_c is proportionally related to T_f (see eq.77). Then the R_{med} becomes almost proportional to the T_c as shown in **Figure 50**. The R_{soc} gradually increase with T_c. This is because the restriction level of C_{soil} would be smaller with increase of T_f.

Effects of EC in soil solution The strontium K_d becomes smaller with the EC in soil solution whereas strontium T_f gradually increases as shown in Figure 30. That is, increase of soil EC prevents the ^{90}Sr transfer from soil to crop. As a result, the R_{soc} and the R_{med} plotted as a function of EC decrease with gentle slopes (**Figure 51**). If the groundwater contamination could be neglected, large

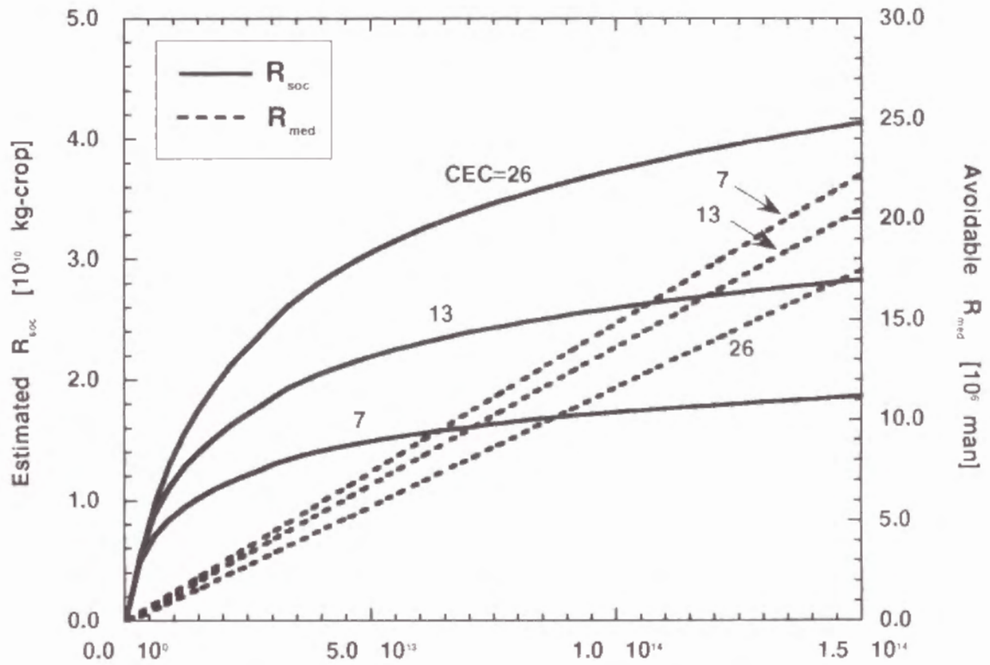

Figure 48. Estimated social risks (R_{soc}s) and avoidable medical risks (R_{med}s) as a function of the amount of ^{90}Sr released from the source (M); the values in the figure are the soil CECs [mmol(+) 100g^{-1}-soil].

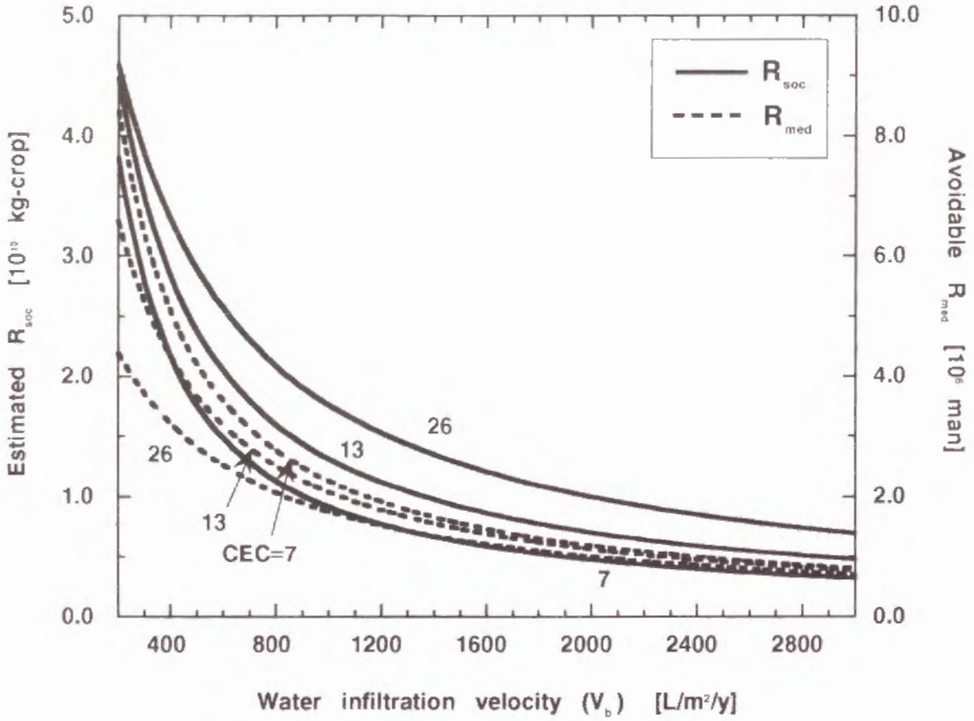

Figure 49. Estimated social risks (R_{soc}s) and avoidable medical risks (R_{med}s) as a function of water infiltration velocity (V_b); the values in the figure are the soil CECs [mmol(+) $100g^{-1}$-soil].

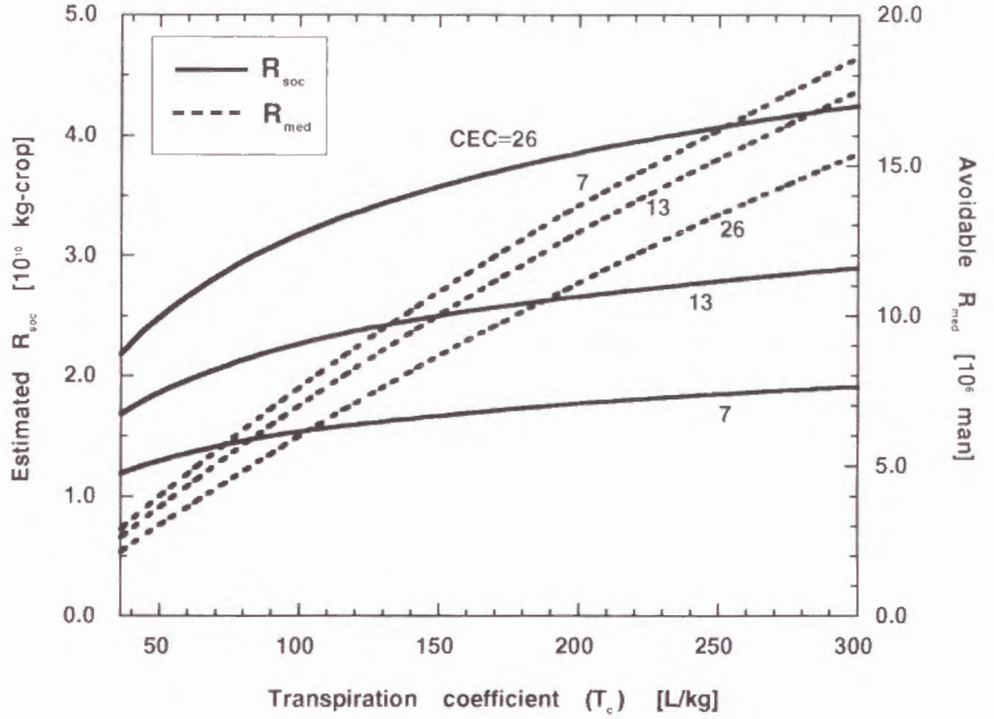

Figure 50. Estimated social risks (R_{soc}s) and avoidable medical risks (R_{med}s) as a function of transpiration coefficient (T_c); the values in the figure are the soil CECs [mmol(+) $100g^{-1}$-soil].

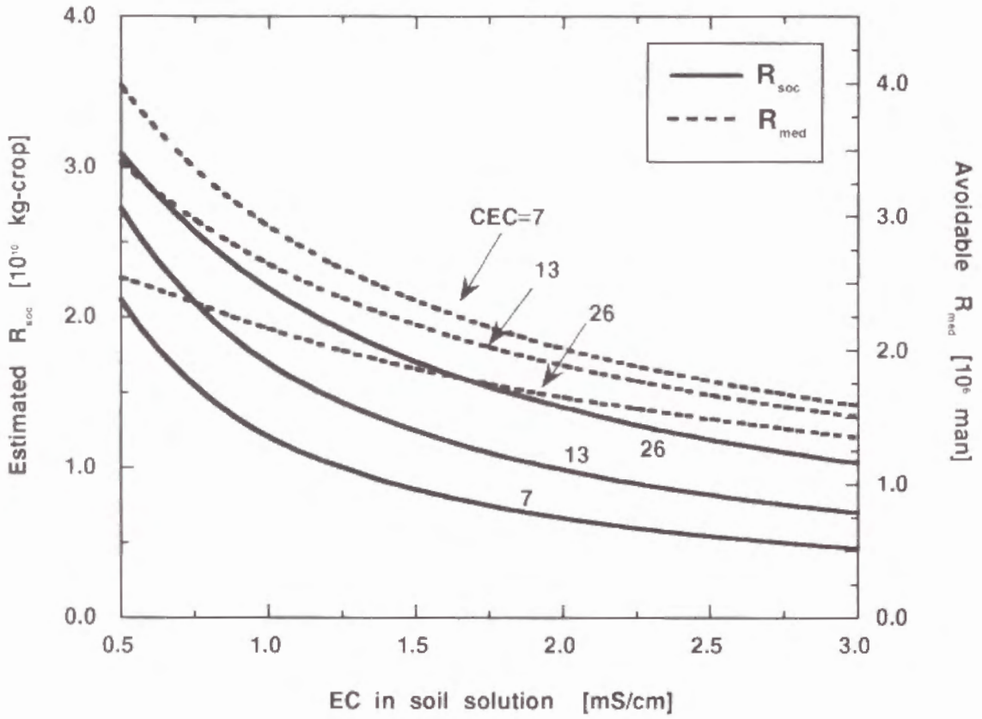

Figure 51. Estimated social risks (R_{soc}s) and avoidable medical risks (R_{med}s) as a function of EC in soil solution; the values in the figure are the soil CECs [mmol(+) $100g^{-1}$-soil].

amounts of irrigation water (high V_b) with high salinity (high EC) would be one of the best countermeasures against the agricultural fields being contaminated by strountium.

5.4.5 Comparison with the method using a constant T_f

As shown in the previous section, the proposed model has a good potential to estimate the social and medical risks realistically by using the information on the growth conditions. Without such information, the reference T_fs as shown in Tables 3 and 4 need to be applied. To prevent underestimation of the dose to the public, some margin is added to T_f. Use of such a constant T_f, however, results in large uncertainty of the estimated risks due to its potential variability caused by the growth condition changes. Hence, this section presents how much error would be produced by the constant T_f in calculated R_{soc} and R_{med} compared to the results by the proposed model.

Here the constant T_f is given as 3.0 which is a ten-fold larger value than the default strontium T_f for vegetable (=0.3) presented by IAEA (1982). The restriction period (Y_R) is determined by eq.84 using this constant T_f and the R_{soc} is calculated from the Y_R. However, the C_{crop} in the R_{med} (eq.86) is calculated by eq.80. The change of the C_{soil} is the same as that calculated by the proposed model as seen in Figure 43. The C_{crop} is predicted by the constant T_f as shown in Figure 44. This figure indicates that the error caused by using the constant T_f is becoming larger for the soil having higher CEC.

The risks estimated by the constant T_f are shown in **Figure 52** as a function of soil CEC with those by the proposed model. Though both R_{soc}s are increasing almost proportionally with the CEC, the slope is larger for the constant T_f than for the model. As a result, the error of the R_{soc} between both methods is becoming larger with the CEC (**Figure 53**). In spite of the large error in the R_{soc}s, the R_{med}s are almost the same even when the CEC is large. This indicates that the increased economic risk (loss of crop yield) when using the constant T_f does not pay for decreasing the health risk. These results suggest that the pertinence of the countermeasure (restricting cultivation) greatly depends on the reliability of the T_f used in the model prediction. It is recommended that soil properties such as soil CEC should be considered in the process of selecting the suitable T_f. The model proposed in this study is expected to be an effective tool for this purpose.

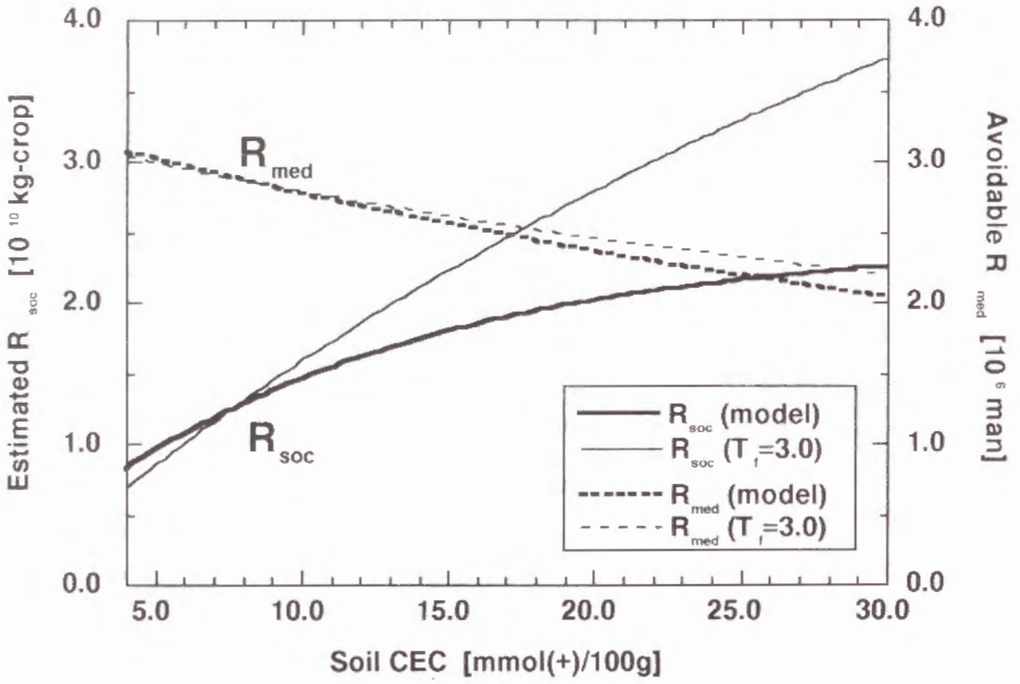

Figure 52. Plots of soil CEC versus avoidable medical risks (R_{med}s) and social risks (R_{soc}s) estimated by a constant T_f (=3.0) and the proposed model.

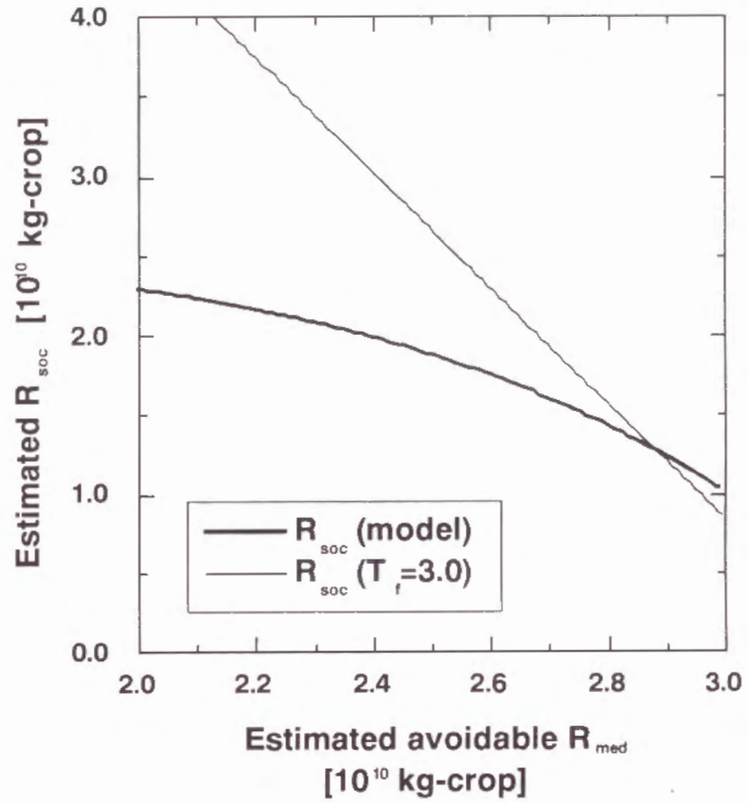

Figure 53. Plots of the social risks (R_{soc}s) estimated by a reference T_f (=3.0) and the proposed model versus the avoidable medical risks (R_{med}s) estimated by the proposed model.

CHAPTER 6. CONCLUSION

6.1. SUMMARY

'How clean is clean ?' is a difficult question to answer. Because the safety level of pollutant concentration should change depending on population, land use, individual human sensitivity (age, sex, and health condition), and information on toxicity and mobility of each pollutant. We, as environmental engineers, are asked to clarify how cleanly the environment should be maintained under consideration of these complicated factors. We should recognize that the authorized Environmental Quality Standards (EQS)are transient values which should be flexible, fitting each situation.

The pertinent assessments on environmental impact of toxic substances cannot be achieved without the help of a reliable environmental transfer model and suitable parameter values. At present, however, a model adapted to the predictions in actual fields has not been sufficiently verified. The parameter values reported have shown large variability but their variational characteristics are still unclear. We need more research to improve the reliability of model predictions.

In Chapter 2, the features, similiarities, and problems of selected well-known transfer models used for environmental impact assessments were briefly reviewed. Among the parameters involved in these models, the reference values of the distribution coefficient (K_d) and the soil-to-plant transfer factor (T_f) were summarized. Both of them do not have sufficient physical meanings; many of the reference values have been obtained empirically without collecting the information as to the growth conditions in spite of the fact that they are greatly affected by various environmental factors. This has caused large uncertainties in the model predictions.

In Chapter 3, the effects of some environmental factors on the radionuclide K_ds (^{54}Mn, ^{60}Co, ^{65}Zn, ^{85}Sr, and ^{137}Cs) were examined by the batch technique to clarify the variation characteristics of the K_ds Thermal effects on the K_ds were observed for all radionuclides. For Mn and Co, the K_ds increased exponentially only at 23 °C in three temperature levels (3°C, 13°C, 23°C); this phenomenon was

considered to be due to biological activities. Increase of competing ion concentrations decreased the K_ds; it was found that the effect of competing ions could be evaluated in a simple way using electrical conductivity (EC). The K_ds obtained for 36 agricultural soils collected throughout Japan showed not normal type distributions but log-normal type distributions. No significant difference between upland and paddy soils was found. Some soil properties had a good correlation with the K_ds of each radionuclide. Among them, the value of soil CEC divided by solution EC, which was assumed to be the cation distribution ratio (CDR), showed an adequate correlation with the K_ds of Sr; the regression equation was K_d=2×CDR.

In Chapter 4, the reference T_fs were statistically analyzed to examine their variational characteristics. Significant differences were found between nuclides, crops, and soil types. It was seen that the statistical information can be utilized to evaluate the suitability of the T_fs adapted for environmental impact assessments. The cause of the variations attributed to soil type was discussed in numerical simulations under different K_ds. The obtained results suggested that the T_f variation would largely depend on the K_d variation even if the meteorological conditions varied. A new model including the K_d for predicting T_f was developed according to the kinetics of root uptake. It consisted of three environmental factors, the selectivity coefficient (S_c), the transpiration coefficient (T_c), and the K_d. This model was validated for Sr in pot experiments with the S_c and the K_d given as functions of the EC in soil solution. The predicted T_f of strontium agreed well with the observations within the error of about 30 %.

Chapter 5 presented new techniques for environmental impact assessments to which the results in the previous chapters were applied. On the basis of statistical information on the K_ds and T_fs obtained in Chapters 3 and 4, the derived intervention levels for surface soil cleanup and for cultivation restriction were determined for the simulated cases of soil contamination using the probabilistic approach. The flexible model developed in Chapter 4 was also applied to a simulated risk assessment. The non-linear relationship observed between the ^{90}Sr concentration in soil (C_{soil}) and that in crop (C_{crop}) was explained theoretically using this model. The results of the sensitivity analyses on the parameters showed that the large sensitivity on both medical and social risks was found in the infiltration

velocity and the soil solution EC. In comparison with the proposed model and the method using a constant T_r, it was suggested that a T_r with a sufficient added margin for safety would bring about a considerable overloading of social risk.

6.2. FUTURE NEEDS

This dissertation has presented some methods to improve the reliability of two empirical parameters, K_d and T_r, both of which are very important and widely used parameters in radiological, environmental impact assessments. These methods presented here are expected to contribute to reliability improvement of environmental impact assessments under selected conditions. Expanding these results to those available in general cases, we need to undertake the following.

- To examine the suitability of the information obtained here for extention to other radionuclides and toxic substances; and, if necessary, to develop practical and reliable methods for the prediction of their behavior.
- To examine the suitability of the information obtained here for extention to other crop species and soil types; and, if necessary, to develop practical and reliable methods applicable to each case.
- To examine the effect of oxidation-reduction potential and microbial activity on the K_ds and T_rs of the nuclides with consideration of their chemical forms.
- To collect other empirical parameter values such as deposition-retention ratio onto crop surface and translocation coefficient to the edible parts and examine their variational characteristics.
- To refine the assessment techniques presented here to make them general and practical tools and, simultaneously, clarify their limitations by considering all the risks caused by soil contamination.

These information will lead to more effective protection of human life from large-scale and long-term environmental pollution which should be a more serious problem for future generations.

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